(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 0 443 059 B1

(12) **EUROPEAN PATENT SPECIFICATION**

(45) Date of publication and mention of the grant of the patent:
17.06.1998 Bulletin 1998/25

(51) Int Cl.[6]: **C07D 231/18**, A01N 43/56, C07F 7/08, C07F 9/6503, C07D 231/22

(21) Application number: **90103338.1**

(22) Date of filing: **21.02.1990**

(54) **3-(Substituted phenyl)pyrazole derivatives, salts thereof, herbicides therefrom, and process for producing said derivatives or salts**

3-(Substituierte Phenyl)pyrazolderivate, deren Salze, deren Herbizide und Verfahren zur Herstellung dieser Derivate oder Salze

Dérivés de 3-(phényl substitué)-pyrazol, leurs sels, leurs herbicides, et les procédés de préparation de ces dérivés ou de leurs sels

(84) Designated Contracting States:
BE ES NL

(43) Date of publication of application:
28.08.1991 Bulletin 1991/35

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(56) References cited:
EP-A- 0 052 333
EP-A- 0 361 114
FR-A- 2 262 663
GB-A- 2 073 172

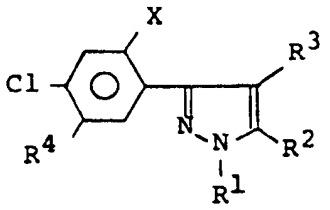
Remarks:
The file contains technical information submitted after the application was filed and not included in this specification

EP 0 443 059 B1

Printed by Jouve, 75001 PARIS (FR)

Description

The present invention relates to 3-(substituted phenyl)pyrazole derivatives or salts thereof, a process for producing said derivatives or salts, and to herbicidal compositions comprising said derivatives or salts and methods for applying said herbicidal compositions. The 3-(substituted phenyl)pyrazole derivatives are represented by the general formula

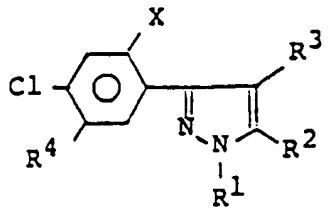


(I)

wherein

X denotes halogen,
R^1 denotes CH_3,
R^2 denotes hydroxy, mercapto, alkylthio, haloalkoxy or haloalkylthio,
R^3 denotes hydrogen or halogen, and
R^4 denotes (i) formyl, (ii) nitro, (iii) -CO-B-R^6 wherein B denotes -O-, -S-, or -NR^7, R^6 and R^7 are the same or different and each denote hydrogen or cycloalkyl, and when B is -O-, R^6 can be an alkali metal atom or -CO-B-R^6 denotes methoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, butoxycarbonyl, methoxycarbonylmethoxycarbonyl, ethoxycarbonylmethoxycarbonyl, propoxycarbonylmethoxycarbonyl, methoxycarbonylethoxycarbonyl, ethoxycarbonylethoxycarbonyl, or propoxycarbonylethoxycarbonyl, (iv) -D-R^8 wherein D denotes -O-,-$S(O)_n$-, n being an integer of 0 to 2, or -NR^9-, R^8 and R^9, which are the same or different, each denotes hydrogen; alkyl; haloalkyl; aminosulfonyl; phenylalkyl or phenoxy-alkyl optionally having on the phenyl ring one or more halogen substituents which are the same or different; or -D-R^8 denotes an alkenyloxy group containing propenyl, butenyl, or pentenyl; an alkynyloxy group containing propynyl,butynyl or pentynyl; an alkenylamino group containing propenyl, butenyl or pentenyl; an alkoxy-carbonylalkoxy or an alkylthiocarbonylalkoxy group containing each methyl, ethyl, propyl or butyl; or (v) -$(CHR^{10})_m$-CO-E-R^{12} wherein E denotes -O-, -S-, or -NR^{11}; R^{10} denotes hydrogen and R^{11} and R^{12}, which are the same or different, each denotes hydrogen; alkyl; haloalkyl; phenyl optionally having on the phenyl ring one or more halogen substituents; phenylalkyl optionally having on the phenyl ring one or more halogen substituents; or R^{11}, jointly with R^{12}, forms a piperidino or morpholinyl group, and when E is -O-, R^{12} can be an alkali metal atom;
and m denotes an integer of 0 to 3.

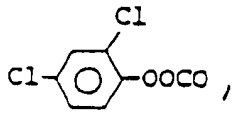
Further, the present invention provides a 3-(substituted phenyl)-pyrazole derivative or a salt thereof, the derivative being represented by the general formula



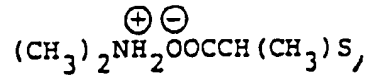
(I)

wherein

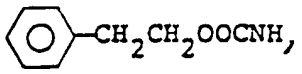
a) X is Cl, R^1 is CH_3, R^2 is CH_3S, R^3 is Cl and R^4 is selected from



$HOOCCH(CH_3)O$, $CH_3OOCCH(CH_3)O$, $C_2H_5OOCCH(CH3)O$, $i\text{-}C_3H_7OOCCH(CH_3)O$, $CH_2{=}CHCH_2OOCCH(CH_3)O$, $CH{\equiv}CCH_2OOCCH(CH_3)O$, $CH_2{=}CHCH_2S$, $CH_2{=}C(Cl)CH_2S$, $CH{\equiv}CCH_2S$, $HO_2CCH(CH_3)S$,



$Cl(CH_2)_3OOCCH(CH_3)S$, $CH_2{=}CHCH_2OOCCH(CH_3)S$, $CH{\equiv}CCH_2OOCCH(CH_3)S$, $C_2H_5CH(CH_3)CH_2OOCCH(CH_3)S$, $(C_2H_5)_2NCOCH(CH_3)S$, CH_3NHSO_2, $(CH_3)_2NSO_2$, $n\text{-}C_3H_7NHSO_2$, $CH_3SO_2NHSO_2$, $Cl^{\ominus}H_3N^{\oplus}$, $CH{\equiv}CCH_2NH$, CH_3OOCNH, CH_3SO_2NH, CH_3NHSO_2NH, $CH_3NHSO_2N(CH_3)$, $(CH_3)_2NSO_2N(CH_3)$, $i\text{-}C_3H_7NHSO_2NH$, $CH{\equiv}CCH_2N(CH_3)SO_2N(CH{\equiv}CCH_2)$, $CH_3OCH_2CH_2OOCNH$, $BrCH_2CH_2OOCNH$, $NCCH_2CH_2OOCNH$, $CH_2{=}C(CH_3)CH_2OOCNH$,



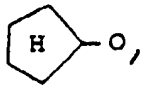
$CH_2{=}CHCH_2OOCCH_2NH$, $CH{\equiv}CCH_2OOCCH_2NH$, $C_2H_5OOCCH(CH_3)NH$, $HOOCCH_2NH$, CH_3OOCCH_2NH, $HOOCCH(CH_3)NH$ and $CH_3OOCCH(CH_3)NH$;

b) X is Cl, R^1 is CH_3, R^2 is CH_3SO, R^3 is Cl and R^4 is selected from NO_2, CH_3SO_2NH, CH_3NHSO_2NH, $i\text{-}C_3H_7NHSO_2NH$ and $CH_3NHSO_2(CH{\equiv}CCH_2)$;

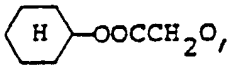
c) X is Cl, R^1 is CHF_2, R^2 is CH_3S, R^3 is Cl and R^4 is selected from NH_2, CH_3NHSO_2NH, $CH_3NHSO_2N(CH{\equiv}CCH_2)$ and $CH_3N(CH{\equiv}CCH_2)SO_2N(CH{\equiv}CCH_2)$;

d) X is Cl, R^1 is CHF_2, R^2 is CH_3SO, R^3 is Cl and R^4 is selected from NH_2 and NO_2;

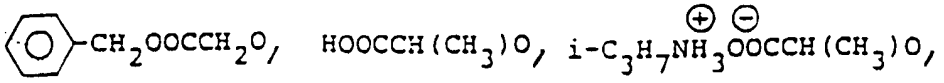
e) X is Cl, R^1 is CH_3, R^2 is CHF_2O, R^3 is Cl and R^4 is selected from



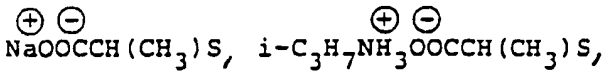
$C_2H_5OCH_2CH_2O$, $NCCH_2O$, CH_3OOCO, $n\text{-}C_6H_{13}OOCCH_2O$, $CH_2{=}CHCH_2OOCCH_2O$, $CH{\equiv}CCH_2OOCCH_2O$,



$C_2H_5O(CH_2)_2OOCCH_2O$, $C_2H_5O(CH_2)_2O(CH_2)_2OOCCH_2O$,

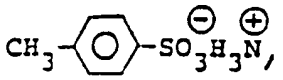


CH_2=$CHCH_2OOCCH(CH_3)O$, $CH{\equiv}CCH_2OOCCH(CH_3)O$, $CH_3NHCOCH(CH_3)O$, $(CH_3)_2NCOCH(CH_3)O$, CH_2-$CHCH_2S$, CH_2=$CHCH_2SO$, $CH{\equiv}CCH_2S$, $(C_2H_5O)_2P(O)CH_2S$, CH_3OOCS, CH_3OOCCH_2S, $C_2H_5OOCCH_2S$, CH_2=$CHCH_2OOCCH_2S$, $HOOCCH(CH_3)S$,

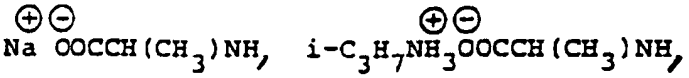


$CH_3OOCCH(CH_3)S$, $C_2H_5OOCCH(CH_3)S$, $C_2H_5OOCCH(CH_3)S(O)$, $C_2H_5OOCCH(CH_3)SO_2$, i-$C_3H_7OOCCH(CH_3)S$, n-$C_4H_9OOCCH(CH_3)S$, $(CH_3)_2CH(CH_2)_2OOCCH(CH_3)S$, CH_2=$CHCH_2OOCCH(CH_3)S$, $CH{\equiv}CCH_2OOCCH(CH_3)S$, $CH_3O(CH_2)_2O(CH_2)_2OOCCH(CH_3)S$, $(C_2H_5O)_2P(O)CH_2OOCCH(CH_3)S$, $CH_3NHCOCH(CH_3)S$, $(CH_3)_2NCOCH(CH_3)S$, $(C_2H_5)_2NCOCH(CH_3)S$, i-$C_3H_7NHCOCH(CH_3)S$,

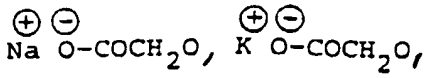
CH_3SO_2NH, CH_3NHSO_2NH, CH_2=$CHCH_2NH$, $CH{\equiv}CCH_2NH$, CH_3OOCNH, $Cl^{\ominus}H_3N^{\oplus}$,

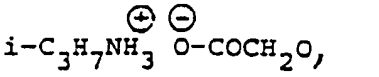


CH_3OOCCH_2NH, $HOOCCH(CH_3)NH$,

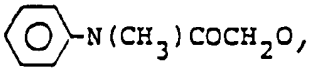


$C_2H_5OOCCH(CH_3)NH$, $CH_3NHOCCH(CH_3)NH$, $(CH_3)_2NOCCH(CH_3)NH$, CH_3NHSO_2, $(CH_3)_2NSO_2$, $(C_2H_5)_2$ NCOS, $(CH_3)_3SiCH_2S$, $(CH_3)_3SiCH_2O$, HO-$COCH_2O$,

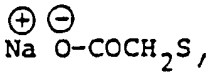


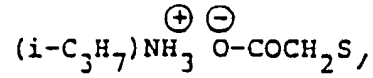


CH_2=$CHC(CH_3)_2O$-$COCH_2O$, $CH_3C{\equiv}CCH_2O$-$COCH_2O$, $CH_3NHCOCH_2O$, $(CH_3)_2NCOCH_2O$, n-C_4H_9 $NHCOCH_2O$, H_2C=$CHCH_2NHCOCH_2O$, $(H_2C{=}CHCH_2)_2NCOCH_2O$,

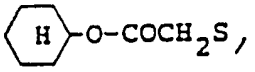


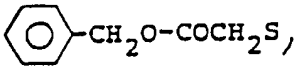
$CH_3SO_2NHCOCH_2O$, $CH_3SO_2NHCOCH(CH_3)O$, $(CH_3)_3SiCH_2O\text{-}COCH_2O$, $CH_3OC_2H_4OC_2H_4O\text{-}COCH(CH_3)O$, $CH_2{=}C(CH_3)CH_2S$, $CH_2{=}C(Cl)CH_2S$, $ClCH{=}CHCH_2S$, $ClCH{=}C(Cl)CH_2S$, $NCCH_2S$,



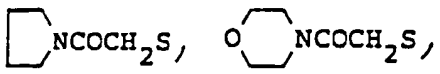


$HO\text{-}COCH_2S$, $i\text{-}C_3H_7O\text{-}COCH_2S$, $n\text{-}C_4H_9O\text{-}COCH_2S$, $C_{12}H_{25}O\text{-}COCH_2S$, $CH_3S\text{-}COCH_2S$, $C_2H_5S\text{-}COCH_2S$, $i\text{-}C_3H_7S\text{-}COCH_2S$, $CH{\equiv}CCH_2O\text{-}COCH_2S$, $C_2H_5OC_2H_4OC_2H_4O\text{-}COCH_2S$,

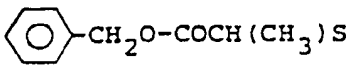




$(CH_3)_3SiCH_2O\text{-}COCH_2S$, $CH_3NHCOCH_2S$, $(CH_3)_2NCOCH_2S$, $H_2C{=}CHCH_2NHCOCH_2S$, $(H_2C{=}CHCH_2)_2NCOCH_2S$, $HC{\equiv}CCH_2NHCOCH_2S$,

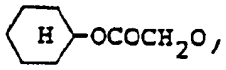


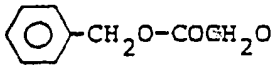
$i\text{-}C_4H_9O\text{-}COCH(CH_3)S$, $t\text{-}C_4H_9CH_2O\text{-}COCH(CH_3)S$, $n\text{-}C_6H_{13}O\text{-}COCH(CH_3)S$,



$C_2H_5S\text{-}COCH(CH_3)S$, $i\text{-}C_3H_7S\text{-}COCH(CH_3)S$, $NCCH(CH_3)O\text{-}COCH(CH_3)S$, $CH_3SC_2H_4O\text{-}COCH(CH_3)S$,

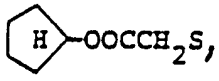
$(CH_3)_3SiCH_2O$-COCH(CH_3)S, CH_2=CHCH_2NHCOCH(CH_3)S, $(H_2C$=CH$CH_2)_2$NCOCH(CH_3)S, HC≡CCH_2NHCOCH(CH_3)S, CH_3CO-O-$(CH_2)_2$S, C_2H_5O-COC$(CH_3)_2$S, CH_3O-COCH(i-C_3H_7)S, C_2H_5O-COCH(CH_3)N(CH_3), CH_2=CHCH_2O-CO, CH_3O-COCH(CH_3)O-CO, C_2H_5O-COCH(CH_3)O-CO, CH≡CCH_2O-CO,



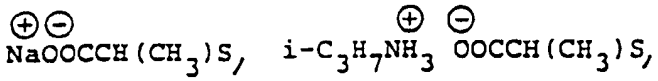


and CH_3O-CO-OCH_2S-CO;

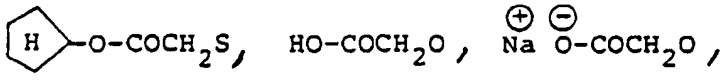
f) X is Cl, R^1 is CH_3, R^2 is CHF_2S, R^3 is Cl and R^4 is selected from CH_2=CHCH_2S, CH≡CCH_2S, $C_2H_5OCH_2CH_2$S, CH_3SCH_2S, NCCH_2S, C_2H_5OOCS, CH_3OOCCH_2S, $C_2H_5OOCCH_2$S,



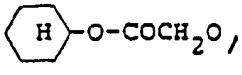
$CH_3NHOCCH_2$S, $(CH_3)_2NOCCH_2$S, HOOCCH(CH_3)S,



$C_2H_5OOCCH(CH_3)$S, n-$C_3H_7OOCCH(CH_3)$S, i-$C_3H_7OOCCH_2$S, $(CH_3)_2CHCH_2OOCCH_2$S, CH_2=C$HCH_2OOCCH(CH_3)$S, $C_2H_5OOC(CH_2)_3$S, $CH_3NHOCCH(CH_3)$S, $(CH_3)_2NOCCH(CH_3)$S, CH≡CCH_2NH, CH_3SO_2NH, CH_3NHSO_2NH, CH_3OOCNH, $(C_2H_5)_2$NCOS, NCCH_2O, ClCH=CHCH_2O, CH_3OCH_2O, $C_2H_5OC_2H_4O$, CH_3OCH_2S, ClCH=CHCH_2S, CH_2=C(Cl)CH_2S,

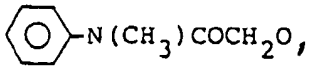


CF_3CH_2O-COCH$_2$O, CH_2=CHCH_2O-COCH$_2$O,



$CH_3NHCOCH_2O$, CH_2=CHCH_2O-COCH(CH_3)O, $CH_3NHCOCH(CH_3)O$, NCCH_2O-COCH$_2$O, CH_3O-COCH(CH_3)

S, $NCCH_2O$-$COCH(CH_3)S$, C_2H_5O-$CO(CH_2)_3S$,

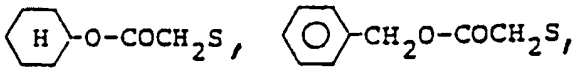


HO-$COCH_2NH$, C_2H_5O-$COCH_2NH$ and C_2H_5O-$COCH(CH_3)NH$;

g) X is F, R^1 is CH_3, R^2 is CH_3SO, R^3 is Cl and R^4 is 5-NO_2;

h) X is F, R^1 is CH_3, R^2 is CH_3S, R^3 is Cl and R^4 is selected from 5-NH_2, 5-SH, 5-$CH{\equiv}CCH_2O$, 5-$CH{\equiv}CCH_2S$ and 5-$C_2H_5OOCCH(CH_3)S$;

i) X is F, R^1 is CH_3, R^2 is CHF_2O, R^3 is Cl and R^4 is selected from HO-$COCH_2O$, CH_3O-$COCH_2O$, $CH_2{=}CHCH_2O$-$COCH_2O$, $CH{\equiv}CCH_2O$-$COCH_2O$, HO-$COCH(CH_3)O$, $CH_2{=}CHCH_2O$-$COCH(CH_3)O$, $CH{\equiv}CCH_2O$-$COCH(CH_3)O$, $CH_2{=}CHCH_2S$, $CH_2{=}C(CH_3)CH_2S$, $CH{\equiv}CCH_2S$, $C_2H_5OC_2H_4S$, HO-$COCH_2S$, CH_3O-$COCH_2S$, C_2H_5O-$COCH_2S$, C_2H_5O-$COCH_2S(O)$, n-C_4H_9O-$COCH_2S$, $CH_2{=}CHCH_2O$-$COCH_2S$, CH_3SCH_2O-$COCH_2S$,



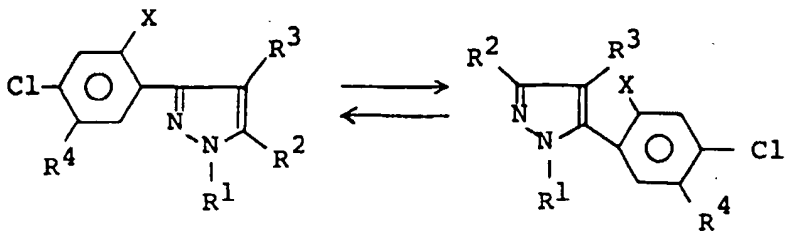
$CH_3NHCOCH_2S$, $(CH_3)_2NCOCH_2S$, HO-$COCH(CH_3)S$, CH_3O-$COCH(CH_3)S$, C_2H_5O-$COCH(CH_3)S$, n-C_4H_9O-$COCH(CH_3)S$, $CH_2{=}CHCH_2O$-$COCH(CH_3)S$, $CH{\equiv}CCH_2O$-$COCH(CH_3)S$, $CH_3OC_2H_4OC_2H_4O$-$COCH(CH_3)S$, CH_3SO_2NH, CH_3NHSO_2NH, C_2H_5O-$COCH_2NH$, C_2H_5O-$COCH(CH_3)NH$, $CH_2{=}CHCH_2O$-CO, $CH{\equiv}CCH_2O$-CO, CH_3O-$COCH(CH_3)O$-CO, C_2H_5O-$COCH(CH_3)O$-CO, $(CH_3)_2NCO$ and

j) X is Cl, R^1 is CH_3, R^2 is CHF_2O, R^3 is Br and R^4 is $BrCH_2CH(Br)CH_2O$ or $BrC{\equiv}CCH_2O$.

The present inventors made intensive studies in order to develop a novel harbicide and as a result have found that 3-(substituted phenyl)pyrazole derivatives represented by the above general formula (I) (hereinafter, simply referred to as formula (I)) and salts thereof are novel compounds, not yet written in literature, and they show excellent herbicidal effects on weeds even at lower dosages. Based on this finding, the present invention has been accomplished.

Prior to the present invention, compounds considered as analogous to the present inventive compounds were disclosed as herbicides in Japanese Patent Application Kokai Nos. Sho.50-117936 (corresponding to GB-A-1488285) Sho.52-91861, Sho.54-70270, and Sho.55-9062 and in other literature. However, the present inventive 3-(substituted phenyl)pyrazole derivative represented by formula (I) or salts thereof have never been disclosed and show superior herbicidal effects at lower dosages than those where the compounds disclosed in the above patent applications do.

The present inventive 3-(substituted phenyl)-pyrazole derivatives represented by formula (I) and salts thereof include structural isomers as shown below. These structural isomers form simultaneously in the production of 3-(substituted phenyl)pyrazole derivatives and each isomer can be isolated by a suitable separating method, e.g. recrystallization or column chromatography. The scope of the present invention also includes these structural isomers.



In this formula, R^1, R^2, R^3, R^4, and X are as defined above.

Certain compounds of the 3-(substituted phenyl)pyrazole derivatives represented by formula (I) have optical isomers, which are also included in the scope of the present invention.

The substituent R^1 in 3-(substituted phenyl)-pyrazole derivatives represented by formula (I) and salts thereof is methyl.

The substituent R^2 can be exemplified by hydroxy, mercapto alkylthio, haloalkoxy, and haloalkylthio groups, of which preferred are haloalkoxy groups and particularly preferred is a difluoromethoxy group.

The substituent R^3 can be exemplified by hydrogen and halogen atoms, of which preferred are halogen atoms, particularly a chlorine atom.

The substituent R^4 can be exemplified by formyl, nitro, -CO-B-R^6 (wherein, B and R^6 are as defined above) and -D-R^8 (wherein, D and R^8 are as defined above). Preferred examples of -CO-B-R^6 are methoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, butoxycarbonyl, methoxycarbonylmethoxycarbonyl, ethoxycarbonylmethoxycarbonyl, propoxycarbonylmethoxycarbonyl, methoxycarbonylethoxycarbonyl, ethoxycarbonylethoxycarbonyl, or propoxycarbonylethoxycarbonyl.

Preferred examples of -D-R^8 are alkoxy and alkylthio groups containing each methyl, ethyl, propyl, or butyl; alkenyloxy groups containing each propenyl, butenyl, or pentenyl; alkynyloxy groups containing each propynyl, butynyl, or pentynyl; alkenylamino groups containing each propenyl, butenyl, or pentenyl; and alkoxy-carbonylalkoxy and alkylthiocarbonylalkoxy groups containing each methyl, ethyl, propyl, or butyl.

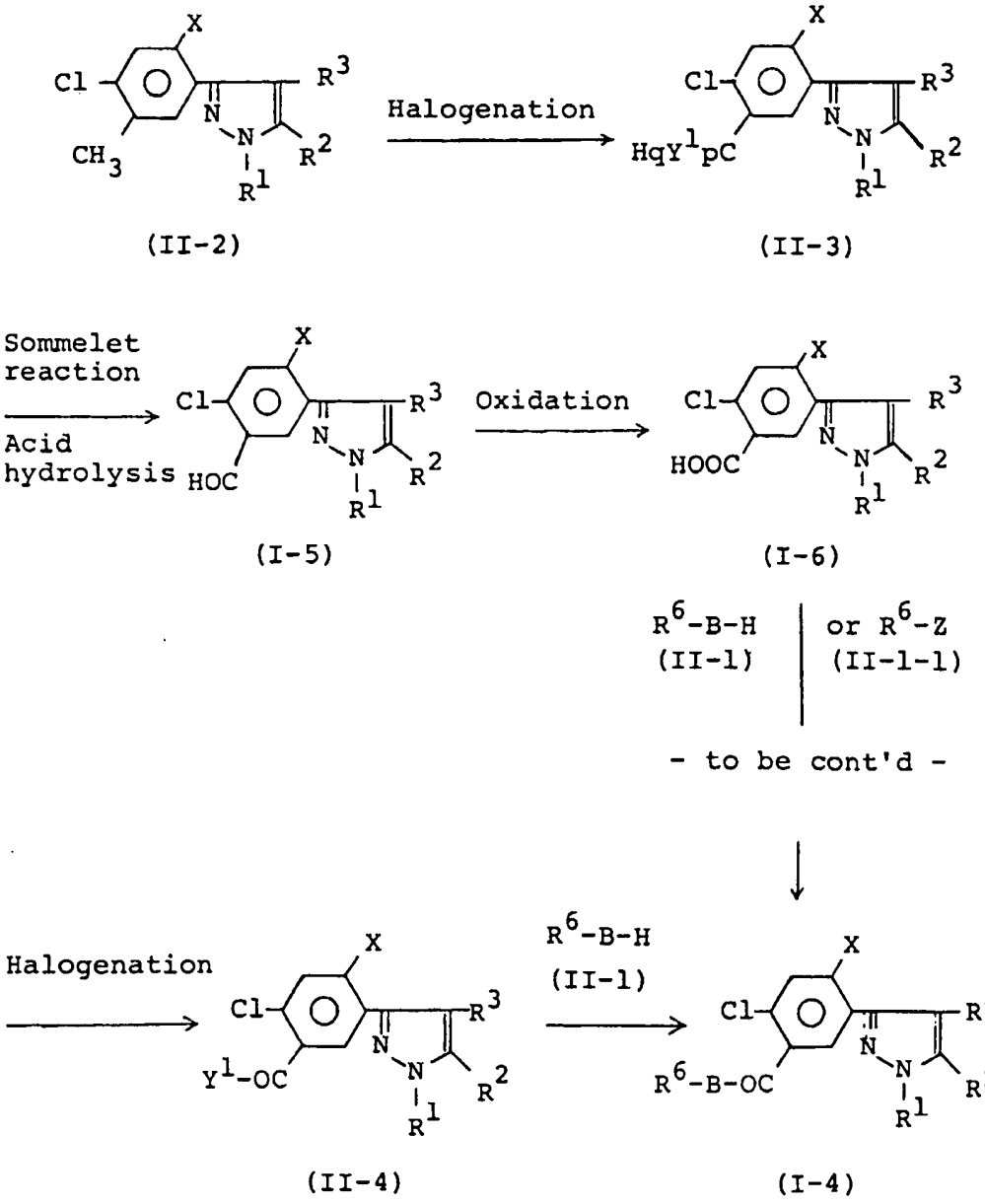
The substituent X is a halogen atom which can be exemplified by chlorine, bromine, fluorine, and iodine, of which preferred are chlorine and fluorine.

Salts of 3-(substituted phenyl)pyrazole derivatives represented by formula (I) are of mineral acids including, e.g. sulfuric acid and hydrochloric acid and of organic acids including, e.g. p-toluenesulfonic acid.

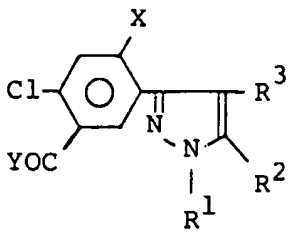
Salts of 3-(substituted phenyl)pyrazole derivatives represented by formula (I) can be produced by treating these derivatives with a suitable mineral acid or organic acid.

Typical processes for producing 3-(substituted phenyl)pyrazole derivatives of formula (I) or salts thereof can be illustrated by the following reaction schemes①, ② and ③

Reaction Scheme (1)



In the above equation, X, R^1, R^2, R^3, and -CO-B-R^6 are as defined above, Y^1 and Z denote halogen, and p and q denote each an integer of 1 or 2 with the proviso that the sum of p and q is 3. That is, the 3-(substituted phenyl)pyrazole derivative represented by formula (I-4) can be produced as follows: A pyrazole derivative represented by formula (II-2) is reacted with a halogenating agent in an inert solvent to give a pyrazole derivative represented by formula (II-3), which in turn is subjected to Sommelet reaction with hexamethylenetetramine in an inert solvent and further hydrolyzed with acid to give a 3-(substituted phenyl)-pyrazole derivative represented by formula (I-5), which in turn is oxidized in an inert solvent to give (I-6), which in turn is halogenated in an inert solvent to give an acid halide represented by formula (II-4). Pyrazole derivatives represented by the following formula (I-3);



(I-3)

in which R^1, R^2, R^3, R^6, X and B are defined above, Y denotes halogen or OH, which consist of a derivative of formula (I-6) (corresponding to the case when Y is OH and a derivative of formula (II-4) (corresponding to the case when Y is halogen) are reacted with a compound represented by formula (II-1) or with a halide represented by formula (II-1-1) in an inert solvent in the presence or absence of a base to produce the 3-(substituted phenyl)pyrazole derivative represented by formula (I-4).

(1) Preparation of compound (II-3) from compound (II-2)

Any inert solvent may be used in this reaction that does not markedly hinder the reaction from proceeding. Such solvents include, though not limited to; halogenated hydrocarbons, e.g. methylene chloride, chloroform, and carbon tetrachloride; aromatic hydrocarbons, e.g. benzene, toluene, and xylene; nitriles, e.g. acetonitrile and benzonitrile; linear ethers, e.g. methyl Cellosolve and diethyl ether; cyclic ethers, e.g. dioxane and tetrahydrofuran; and sulforane, dimethyl sulfone and dimethylsulfoxide. These solvents may be used alone or in combination.

Halogenating agents suitable for use in the above reaction include, for example, N-bromosuccinimide (NBS), N-chlorosuccinimide (NCS), bromine, chlorine, t-butyl hypohalogenite, and trichloromethanesulfonyl halide. When NBS or NCS is used as a halogenating agent, it is preferable to use an organic peroxide such as benzoyl peroxide or light as a catalyst or exciter.

The amount of halogenating agent used may be chosen from the range of 1 mole to an excess of 1 mole per mole of the pyrazole derivative of formula (II-2).

The reaction temperature ranges from 0°C to the boiling point of the inert solvent used.

The reaction period is chosen from the range of several minutes to 48 hours so as to complete the reaction.

After completion of the reaction, the 3-(substituted phenyl)pyrazole derivative of formula (II-3) can be obtained from the reaction product solution by an ordinary method, e.g. solvent extraction, and if necessary, by purification procedure such as recrystallization or column chromatography.

(2) Preparation of compound (I-5) from compound (II-3)

This reaction process consists of the first step of carrying out Sommelet reaction and the second step of carrying out acid hydrolysis. Inert solvents suitable for use in the first step include organic acids, e.g. glacial acetic acid, mineral acids, e.g. hydrochloric acid and sulfuric acid, water, and mixtures of these solvents.

Hexamethylenetetramine is used in an amount equimolar to the pyrazole derivative of formula (II-3). An excess of hexamethylenetetramine can also be used.

The reaction temperature ranges from 10°C to the boiling point of inert solvent used, preferably from 30 to 180°C.

After completion of the reaction, the resultant solution containing the objective compound is subjected to the acid hydrolysis in the second step.

To the acid hydrolysis, the reaction product solution from the first step is subjected as such or with a large excess of mineral acid such as hydrochloric acid or sulfuric acid added as occasion demands, the latter procedure being preferable.

The reaction temperature is in the range of 80 to 180°C and the reaction period is chosen from the range of several minutes to 48 hours so as to complete the reaction.

After completion of the reaction, the 3-(substituted phenyl)pyrazole derivative of formula (I-5) can be obtained from the reaction product solution by an ordinary method, e.g. solvent extraction, and if necessary, by purification procedure such as recrystallization or column chromatography.

(3) Preparation of compound (I-6) from compound (I-5)

Inert solvents suitable for use in this reaction include aromatic hydrocarbons such as benzene, toluene, and xylene, and further pyridine and its derivatives and water. These solvents may be used alone or in combination.

Oxidants, e.g. potassium permanganate and potasium dichromate, can be used as oxidizing agents in amounts of 1 to 5 moles, preferably 1 to 2 moles, per mole of the 3-(substituted phenyl)pyrazole derivative of formula (I-5).

The reaction temperature is from 0°C to the boiling point of inert solvent used, preferably in the range of 30 to 180°C. The reaction period is chosen from the range of several minutes to 48 hours so as to complete the reaction.

After completion of the reaction, the 3-(substituted phenyl)pyrazole derivative of formula (I-6) can be obtained from the reaction product solution by an ordinary method e.g. solvent extraction, and if necessary, by purification procedure such as recrystallization or column chromatography.

(4) Preparation of compound (II-4) from compound (I-6)

Inert solvents suitable for use in this acid halide-forming reaction include; halogenated hydrocarbons, e.g. methylene chloride, chloroform, and carbon tetrachloride; aromatic hydrocarbons, e.g. benzene, toluene, and xylene; linear ethers, e.g. methyl Cellosolve, diethyl ether, and diisopropyl ether; and cyclic ethers, e.g. dioxane and tetrahydrofuran.

Halogenating agents suitable for this reaction include, for example, thionyl chloride, phosphorus pentachloride, and phosphorus trichloride.

The halogenating agent may be used in amounts of 1 mole to an excess of 1 mole, preferably in such an excess, per mole of the 3-(substituted phenyl)pyrazole derivative of formula (I-6).

A catalytic amount of triethylamine, pyridine, dimethylformamide, or the like may be added for the purpose of promoting this reaction.

The reaction temperature is in the range of room temperature to the boiling point of solvent used. The reaction period ranges from several minutes to 48 hours depending upon the amounts of reactants used and the reaction temperature.

After completion of the reaction, the excess halogenating agent and the solvent are removed from the reaction product solution by evaporation to isolate the objective compound, which may be subjected to the next reaction after purification, if necessary, by recrystallization, distillation, or some other method.

(5) Preparation of compound (I-4) from compound (I-6) or from compound (II-4)

This reaction is esterification or amide-forming reaction. The esterification can be carried out by using an excess of the alcohol corresponding to the objective ester in the presence of a mineral acid, e.g. concentrated sulfuric acid, which acts as a reactant and a solvent. For the esterification, the 3-(substituted phenyl)pyrazole derivative of formula (I-6) can also be used in the form of alkali metal salt.

The esterification or the amide-forming reaction can also be carried out in the presence of an inert solvent and a dehalogenating agent. Inert solvents suitable for use in the acid halide-forming reaction can also be used in this case.

The alcohol or amine or the halide of formula (II-1-1) may be used in an equimolar amount or more.

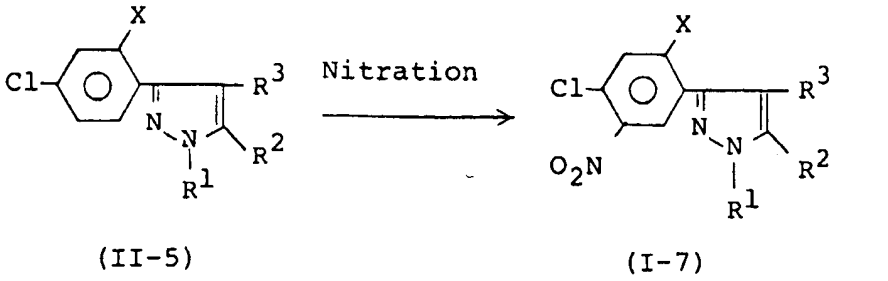
Dehalogenating agents suitable for use in this reaction are inorganic bases and organic bases. The inorganic bases include alkali metal salts, e.g. sodium hydroxide and potassium hydroxide and the organic bases include tertiary amines, e.g. triethylamine, and 4-dimethylaminopyridine, and 1,8-diazabicyclo-[5,4,0]-7-undecene (DBU).

The reaction temperature is in the range of 0°C to the boiling point of the solvent used, preferably in the range of 0 to 150°C. The reaction period ranges from several minutes to 48 hours depending upon the amounts of reactants used, the reaction temperature, etc.

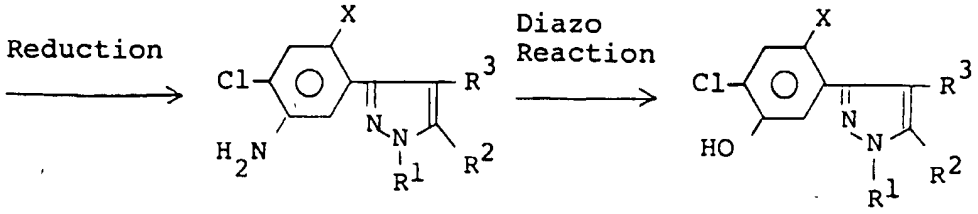
After completion of the reaction, the 3-(substituted phenyl)pyrazole derivative of formula (I-4) can be obtained from the reaction product solution by an ordinary method, e.g. solvent extraction, and if necessary, by purification procedure such as recrystallization or column chromatography.

Reaction Scheme (2)

Process A

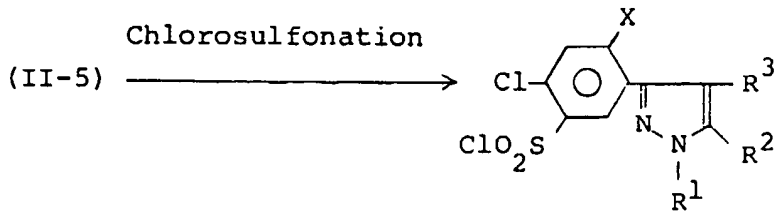


(II-5) (I-7)

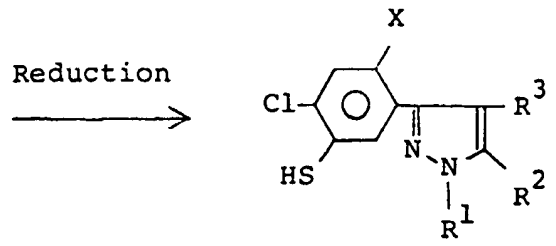


(I-8) (I-9)

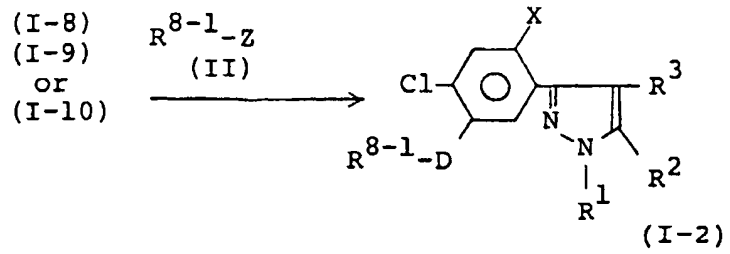
Process B



(II-6)



(I-10)

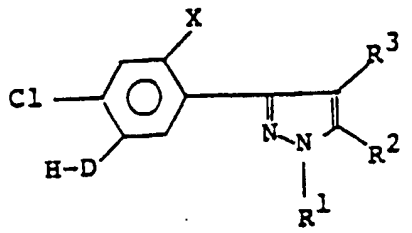


(I-2)

In the above equations, R^1, R^2, R^3, R^{8-1}, D, X, and Z are as defined above.

That is, a pyrazole derivative of formula (II-5) is subjected to nitration with a nitrating agent in an inert solvent to give a 3-(substituted phenyl)-pyrazole derivative of formula (I-7). This derivative is then reduced in an inert solvent to give a 3-(substituted phenyl)pyrazole derivative of formula (I-8), which is further diazotized in an inert solvent and then decomposed, whereby a 3-(substituted phenyl)-pyrazole derivative of formula (I-9) can be obtained. On the other hand, a pyrazole derivative of formula (II-5) is subjected to chlorosulfonyl substitution in an inert solvent to give a pyrazole derivative of formula (II-6), which in turn is reduced, whereby a 3-(substituted phenyl)pyrazole derivative of formula (I-10) can be obtained.

Further, a 3- (substituted phenyl)pyrazole derivative of formula (I-2) can be produced from a 3- (substituted phenyl) pyrozole derivative represented by the general formula



(I-1)

wherein X, R^1, R^2, R^3, and D are as defined above, especially of formula (I-8), (I-9), or (I-10) by reacting with a halide of formula (II) in an inert solvent in the presence of a base.

When R^{8-1} in the 3-(substituted phenyl)-pyrazole derivative of formula (I-2) is $-CH\cdot R^{10}-CO\cdot E\cdot R^{12}$ (wherein, R^{10}, R^{12}, and E are as defined above), this 3-(substituted phenyl)pyrazole derivative can be produced by converting an acid represented by $-CHR^{10}-CO-E-H$ to an acid halide in the same manner as in the above reaction process ① and subjecting the acid halide to esterification, thiol esterification, or amide-forming reaction.

(1) Preparation of compound (I-7) from compound (II-5)

Nitrating agents suitable for use in this reaction include concentrated nitric acid, a mixture of fuming nitric acid with concentrated sulfuric acid, and acetyl nitrate that is formed by mixing concentrated nitric acid with acetic anhydride.

Mineral acids such as sulfuric acid and hydrochloric acid can be used as solvents in this process.

Since this reaction is an equimolar reaction, the use of an equimolar amount of nitrating agent is sufficient though

it may be used in excess.

The reaction temperature is in the range of -10 to 140°C, preferably -10 to 20°C. The reaction period is chosen from the range of several minutes to 48 hours depending upon the amounts of reactants charged, the reaction temperature, etc.

After completion of the reaction, the intended 3-(substituted-phenyl)pyrazole derivative of formula (I-7) can be obtained from the reaction product solution by either pouring it into ice-cold water and collecting the precipitated crystals or subjecting the solution to solvent extraction or other separating procedure to isolate the intended product, and if necessary, by further purification procedure such as recrystallization.

(2) Preparation compound (I-8) from compound (I-7)

Any inert solvent that does not markedly hinder the reaction from proceeding may be used in this reaction. Such inert solvents include; alcohols, e.g. methanol, ethanol, and propanol; linear ethers, e.g. diethyl ether and methyl Cellosolve; cyclic ethers, e.g. dioxane and tetrahydrofuran; organic acids, e.g. acetic acid; mineral acids, e.g. hydrochloric acid; and water.

Reducing agents suitable for this reaction in an acidic medium include, for example, zinc, iorn, tin, and tin chloride. Zinc dust can be used in a neutral or basic medium. For the catalytic hydrogenation, which can be carried out under normal or elevated pressure, suitable catalysts include, for example, Raney nickel, palladium-carbon, palladium oxide, platinum, platinum black, platinum sulfide-carbon, and rhodium-alumina.

In an acidic medium, the reducing agent may be used in an equimolar amount though generally in excess. For the catalytic hydrogenation, the amount of Raney nickel or the like when it is used is from 5 to 20% by weight based on the weight of the 3-(substituted)pyrazole derivative of formula (I-7) and the amount of noble metal such as platinum or palladium when it is used is from 0.02 to 5% by weight on the the same basis.

The reaction temperature is in the range of 0 to 150°C, preferably 10 to 100°C. The reaction period may be chosen from the range of several minutes to 48 hours depending upon the amounts of reactants charged, the reaction temperature, etc.

After completion of the reaction, the intended 3-(substituted phenyl)pyrazole derivative of formula (I-8) can be obtained in the following manner: When the reaction is carried out in an acidic medium, the reaction product solution is poured into ice-cold water, this mixture is turned basic, and the objective product is isolated by a method such as solvent extraction. In case of catalytic hydrogenation, the catalyst is removed from the reaction product fluid by filtration and the objective product is recovered by concentrating the filtrate. If necessary, the objective product is further purified by recrystallization, column chromatography, or some other method.

(3) Preparation of compound (I-9) from compound (I-8)

This process consists of the step of diazotization and the step of decomposition. In the diazotization step, a mineral acid such as sulfuric acid or hydrochloric acid can be used as an inert solvent. For the diazotization, sodium nitrite in the form of powder or aqueous solution of suitable concentration is used in an equimolar amount or in excess.

The reaction temperature is in the range of -5 to 5°C.

The resulting diazonium salt is subjected to decomposition without isolation.

In the decomposition step, 30 to 70% sulfuric acid may be used as an inert solvent.

The decomposition is carried out in the presence of a catalyst at a temperature of 10 to 180°C. When the diazonium salt in solution form is added dropwise to aqueous sulfuric acid, the reaction is conducted at a temperature of 50 to 180°C. When cuprous oxide is used, as a catalyst, the reaction temperature is in the range of 10 to 50°C.

Copper nitrate and cuprous oxide can be used as catalysts. To the reaction system is added copper nitrate and then cuprous oxide is added in limited amounts.

Copper nitrate is used in an amount of 1 to 60 moles, preferably 30 to 60 moles, and cuprous oxide is used in an amount of catalytic to 2 moles, preferably about 1 mole, per mole of the diazonium salt.

The total reaction period of diazotization and decomposition may be chosen from the range of several minutes to 48 hours depending upon the amounts of reactants charged and the reaction temperature.

After completion of the reaction, the 3-(substituted phenyl)pyrazole derivative of formula (I-9) can be obtained by pouring the reaction product solution into ice-cold water, isolating the objective product from the mixture by a method such as solvent extraction, and if necessary, purifying the isolated product by recrystallization, column chromatography, or some other method.

(4) Preparation of compound (II-6) from compound (II-5)

Any solvent may be used for this reaction that does not markedly hinder the reaction. Such solvents include;

halogenated hydrocarbons, e.g. methylene chloride, chloroform, and carbon tetrachloride, aliphatic nitriles, e.g. acetonitrile; linear ethers, e.g. methyl Cellosolve; cyclic ethers, e.g. dioxane and tetrahydrofuran; and mineral acids, e.g. concentrated sulfuric acid.

For this chlorosulfonyl substitution, chlorosulfonic acid or the like may be used. Another available method comprises sulfonating the starting compound of formula (II-5) with fuming sulfuric acid, converting the resulting sulfonic acid to an alkali metal salt, and chlorinating it with phosphorus pentachloride to give the objective product. A further method comprises reacting the starting compound with fuming sulfuric acid, followed by the reaction with carbon tetrachloride to give the objective product.

Chlorosulfonic acid is used in an equimolar amount or in excess, preferably in excess.

Fuming sulfuric acid is used also in an equimolar amount or in excess, preferably in excess. Carbon tetrachloride is used in an amount chosen properly from the range of an equimolar amount to an excess.

The reaction temperature is in the range of 0 to 180°C, preferably 15 to 100°C. The reaction period may be chosen from the range of several minutes to 48 hours depending upon the amounts of reactants charged and the reaction temperature.

After completion of the reaction, the 3-(substituted phenyl)pyrazole derivative of formula (II-6) can be obtained by pouring the reaction product solution into water, isolating the objective product from the resulting mixture by solvent extraction, and if necessary, purifying the isolated product by recrystallization, column chromatography, or some other procedure.

(5) Preparation of compound (I-10) from compound (II-6)

This reduction can be carried out in an inert solvent such as glacial acetic acid. Reducing agents suitable for this reaction include, for example, zinc, tin, and tin chloride, which may be used in an amount of 1 mole or more, preferably 5 moles or more, per mole of the compound of formula (II-6).

The reaction temperature is in the range of 0 to 180°C, preferably 15 to 120°C. The reaction period is chosen from the range of several minutes to 48 hours depending upon the amounts of reactants charged and the reaction temperature.

After completion of the reaction, the 3-(substituted phenyl)pyrazole derivative of formula (I-10) can be obtained by pouring the reaction product solution into water, isolating the objective product from the mixture by solvent extraction, and if necessary, purifying the isolated product by recrystallization, column chromatography, or some other procedure.

(6) Preparation of compound (I-2) from compound (I-9) or compound (I-10)

This reaction can be carried out according to the procedure of the above reaction process ① 5 , whereby the 3-(substituted phenyl)pyrazole derivative of formula (I-2) can be produced.

③ Salts of 3-(substituted phenyl)pyrazole derivatives of formula (I)

These salts are formed from acids including mineral acids, e.g. hydrochloric acid and sulfuric acid, and organic acids, e.g. p-toluenesulfonic acid. The production of these salts can be carried out by treating 3-(substituted phenyl) pyrazole derivatives of formula (I), obtained according to the above stated process, with the above-cited mineral acid or organic acid.

Typical examples of the 3-(substituted phenyl)pyrazole derivatives represented by formula (I) and the salts thereof are shown in Table 1.

Formula (I):

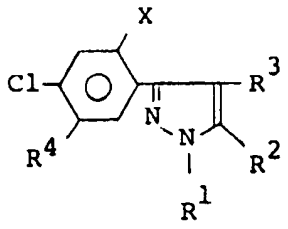
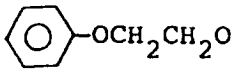
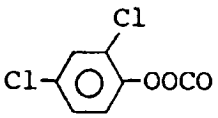
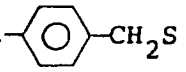


Table 1

Comp'd No.	R^1	R^2	R^3	R^4	X	Property
1	CH_3	CH_3S	H	HO	Cl	m.p. 157.1°C
2	CH_3	CH_3S	Cl	HO	Cl	m.p. 163.2°C
3	CH_3	CH_3S	Cl	$i\text{-}C_3H_7O$	Cl	nD 1.5882 (26.5°C)
4	CH_3	CH_3S	Cl	$CH_2{=}CHCH_2O$	Cl	nD 1.6131 (25.3°C)
5	CH_3	CH_3S	Cl	$CH{\equiv}CCH_2O$	Cl	m.p. 71.5 - 72.5°C
6	CH_3	CH_3S	Cl	$C_2H_5OCH_2CH_2O$	Cl	nD 1.5944 (25,8°C)
7	CH_3	CH_3S	Cl		Cl	nD 1.6075 (25.6°C)
8	CH_3	CH_3S	Cl		Cl	nD 1.5988 (25.9°C)
9	CH_3	CH_3S	Cl	$HOOCCH(CH_3)O$	Cl	m.p. 191 - 194°C
10	CH_3	CH_3S	Cl	$CH_3OOCCH(CH_3)O$	Cl	m.p. 90 - 93°C

- cont'd -

11	CH_3	CH_3S	Cl	$C_2H_5OOCCH(CH_3)O$	Cl	nD 1.5763(28.8°C)
12	CH_3	CH_3S	Cl	$i\text{-}C_3H_7OOCCH(CH_3)O$	Cl	m.p. 87 - 90°C
13	CH_3	CH_3S	Cl	$CH_2{=}CHCH_2OOCCH(CH_3)O$	Cl	nD 1.5802(23.4°C)
14	CH_3	CH_3S	Cl	$CH{\equiv}CCH_2OOCCH(CH_3)O$	Cl	nD 1.5858(23.7°C)
15	CH_3	CH_3S	Cl	HS	Cl	nD 1.6303(24.3°C)
16	CH_3	CH_3S	Cl	$i\text{-}C_3H_7S$	Cl	m.p. 91.8°C
17	CH_3	CH_3S	Cl	CHF_2S	Cl	m.p. 87.2°C
18	CH_3	CH_3S	Cl	CHF_2CF_2S	Cl	nD 1.5820(24.7°C)
19	CH_3	CH_3S	Cl	$CH_2{=}\ CHCH_2S$	Cl	Viscous substance
20	CH_3	CH_3S	Cl	$CH_2{=}C(Cl)CH_2S$	Cl	nD 1.6370(25.1°C)
21	CH_3	CH_3S	Cl	$CH{\equiv}CCH_2S$	Cl	m.p. 94.5°C
22	CH_3	CH_3S	Cl		Cl	nD 1.6528(24.9°C)
23	CH_3	CH_3S	Cl	$C_2H_5OOCCH_2S$	Cl	m.p. 112.1°C
24	CH_3	CH_3S	Cl	$C_2H_5OOCCH_2CH_2S$	Cl	m.p. 74.8°C
25	CH_3	CH_3S	Cl	$HO_2CCH(CH_3)S$	Cl	m.p. 180.0°C

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26	CH_3	CH_3S	Cl	$i\text{-}C_3H_7\overset{\oplus}{N}H_3\overset{\ominus}{O}OCCH(CH_3)S$	Cl	nD 1.5878(22.1°C)
27	CH_3	CH_3S	Cl	$(CH_3)_2\overset{\oplus}{N}H_2\overset{\ominus}{O}OCCH(CH_3)S$	Cl	m.p. 176.1°C
28	CH_3	CH_3S	Cl	$CH_3OOCCH(CH_3)S$	Cl	nD 1.6131(24.9°C)
29	CH_3	CH_3S	Cl	$C_2H_5OOCCH(CH_3)S$	Cl	nD 1.6010(25.0°C)
30	CH_3	CH_3S	Cl	$i\text{-}C_3H_7OOCCH(CH_3)S$	Cl	nD 1.5936(25.0°C)
31	CH_3	CH_3S	Cl	$Cl(CH_2)_3OOCCH(CH_3)S$	Cl	nD 1.6025(25.2°C)
32	CH_3	CH_3S	Cl	$CH_2{=}CHCH_2OOCCH(CH_3)S$	Cl	nD 1.6030(25.0°C)
33	CH_3	CH_3S	Cl	$CH{\equiv}CCH_2OOCCH(CH_3)S$	Cl	nD 1.5954(25.0°C)
34	CH_3	CH_3S	Cl	$C_2H_5CH(CH_3)CH_2OOCCH(CH_3)S$	Cl	nD 1.5834(25.0°C)
35	CH_3	CH_3S	Cl	$(C_2H_5)_2NCOCH(CH_3)S$	Cl	nD 1.5983(26.4°C)
36	CH_3	CH_3S	Cl	H_2NSO_2	Cl	m.p. 203.5°C
37	CH_3	CH_3S	Cl	CH_3NHSO_2	Cl	m.p. 79.1°C
38	CH_3	CH_3S	Cl	$(CH_3)_2NSO_2$	Cl	m.p. 135.7°C
39	CH_3	CH_3S	Cl	$n\text{-}C_3H_7NHSO_2$	Cl	m.p. 102-105°C
40	CH_3	CH_3S	Cl	$CH_3SO_2NHSO_2$	Cl	m.p. 184 - 187°C

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41	CH_3	CH_3S	Cl	H_2N	Cl	nD 1.6403(25.0°C)
42	"	"	"	$Cl^{\ominus}H_3N^{\oplus}$	"	m.p. 174.5°C
43	"	"	"	CH_3NH	"	nD 1.6368(21.7°C)
44	"	"	"	$(CH_3)_2N$	"	nD 1.6241(22.1°C)
45	"	"	"	$CH_2=CHCH_2NH$	"	nD 1.6238(22.2°C)
46	"	"	"	$(CH_2=CHCH_2)_2N$	"	nD 1.6088(21.6°C)
47	"	"	"	$CH\equiv CCH_2NH$	"	m.p. 140.8°C
48	"	"	"	CH_3OOCNH	"	nD 1.6301(22.2°C)
49	"	"	"	HOOC	"	m.p. 213 - 214°C
50	"	"	"	C_2H_5OOC	"	nD 1.6029(20.1°C)
51	"	"	"	CH_3SO_2NH	"	nD 1.6002(25.9°C)
52	"	"	"	CH_3NHSO_2NH	"	m.p. 134.5°C
53	"	"	"	$CH_3NHSO_2N(CH_3)$	"	nD 1.5763(26.4°C)
54	"	"	"	$(CH_3)_2NSO_2N(CH_3)$	"	nD 1.5839(26.0°C)
55	"	"	"	$i\text{-}C_3H_7NHSO_2NH$	"	m.p. 148.6°C

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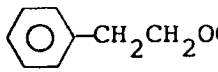
56	CH_3	CH_3S	Cl	$CH{\equiv}CCH_2N(CH_3)SO_2N(CH{\equiv}CCH_2)$	Cl	m.p. 111.5°C
57	"	"	"	$CH_3OCH_2CH_2OOCNH$	"	m.p. 112 - 115°C
58	"	"	"	$BrCH_2CH_2OOCNH$	"	m.p. 119.5°C
59	"	"	"	$NCCH_2CH_2OOCNH$	"	nD 1.5938(24.7°C)
60	"	"	"	$CH_2{=}C(CH_3)CH_2OOCNH$	"	m.p. 125.8°C
61	"	"	"		"	m.p. 103 - 105°C
62	"	"	"	$CH_2{=}CHCH_2OOCCH_2NH$	"	m.p. 112.0°C
63	"	"	"	$CH{\equiv}CCH_2OOCCH_2NH$	"	m.p. 127.0°C
64	"	"	"	$C_2H_5OOCCH(CH_3)NH$	"	Viscous substance
					"	
66	"	"	"	$HOOCCH_2NH$	"	m.p. 207.1°C
67	"	"	"	CH_3OOCCH_2NH	"	m.p. 132.4°C
68	"	"	"	$HOOCCH(CH_3)NH$	"	m.p. 218.7°C
69	"	"	"	$CH_3OOCCH(CH_3)NH$	"	m.p. 144.2°C

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70	CH_3	CH_3SO	Cl	NO_2	Cl	m.p. 168.1°C
71	"	"	"	CH_3SO_2NH	"	m.p. 168.0°C
72	"	"	"	CH_3NHSO_2NH	"	m.p. 146.3°C
73	"	"	"	$i\text{-}C_3H_7NHSO_2NH$	"	m.p. 184.8°C
74	"	"	"	$CH_3NHSO_2N(CH{\equiv}CCH_2)$	"	m.p. 192.5°C
75	CHF_2	CH_3S	"	NH_2	"	m.p. 87.4°C
76	"	"	"	CH_3NHSO_2NH	"	m.p. 133.3°C
77	"	"	"	$CH_3NHSO_2N(CH{\equiv}CCH_2)$	"	nD 1.5625(25.6°C)
78	"	"	"	$CH_3N(CH{\equiv}CCH_2)SO_2N(CH{\equiv}CCH_2)$	"	nD 1.5675(25.6°C)
79	"	CH_3SO	"	NH_2	"	m.p. 187.1°C
80	"	"	"	NO_2	"	m.p. 165.1°C
81	CH_3	CHF_2O	"	CH_3O	"	nD 1.5567(23.4°C)
82	"	"	"	C_2H_5O	"	nD 1.5434(23.5°C)
83	"	"	"	$n\text{-}C_3H_7O$	"	nD 1.5410(25.1°C)
84	"	"	"	$i\text{-}C_3H_7O$	"	nD 1.5422(23.5°C)

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	CH_3	CHF_2O	Cl		Cl	nD
85	"	"	"	$CH_2=CHCH_2O$	"	nD 1.5482(25.4°C)
86	"	"	"	$CH_2=CHCH_2O$	"	nD 1.5536(28.4°C)
87	"	"	"	$CH\equiv CCH_2O$	"	m.p. 84.0°C
88	"	"	"	$C_2H_5OCH_2CH_2O$	"	m.p. 79.5°C
89	"	"	"	$NCCH_2O$	"	m.p. 98.9°C
90	"	"	"	CH_3OCO	"	nD 1.5387(23.2°C)
91	"	"	"	CH_3OCCH_2O	"	m.p. 119.8°C
92	"	"	"	$n\text{-}C_3H_7OCCH_2O$	"	m.p. 89.7°C
93	"	"	"	$i\text{-}C_3H_7OCCH_2O$	"	m.p. 105.0°C
94	"	"	"	$n\text{-}C_4H_9OCCH_2O$	"	m.p. 75.3°C
95	"	"	"	$CH_3CH_2CH(CH_3)OCCH_2O$	"	m.p. 107.8°C
96	"	"	"	$n\text{-}C_6H_{13}OCCH_2O$	"	nD 1.5244(23.0°C)
97	"	"	"	$CH_2=CHCH_2OCCH_2O$	"	m.p. 84.7°C
98	"	"	"	$CH\equiv CCH_2OCCH_2O$	"	m.p. 119.6°C

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99	CH_3	CHF_2O	Cl		Cl	m.p. 91.8°C
100	"	"	"	$C_2H_5O(CH_2)_2OOCCH_2O$	"	nD 1.5261(22.9°C)
101	"	"	"	$C_2H_5O(CH_2)_2O(CH_2)_2OOCCH_2O$	"	nD 1.5287(23.0°C)
102	"	"	"		"	m.p. 139.1°C
103	"	"	"	$HOOCCH(CH_3)O$	"	m.p. 158.7°C
104	"	"	"	$i\text{-}C_3H_7\overset{\oplus}{N}H_3\overset{\ominus}{O}OCCH(CH_3)O$	"	Viscous substance
105	"	"	"	$C_2H_5OOCCH(CH_3)O$	"	nD 1.5238(25.7°C)
106	"	"	"	$n\text{-}C_4H_9OOCCH(CH_3)O$	"	nD 1.5253(18.5°C)
107	"	"	"	$CH_2=CHCH_2OOCCH(CH_3)O$	"	m.p. 84.3°C
108	"	"	"	$CH\equiv CCH_2OOCCH(CH_3)O$	"	m.p. 109.7°C
109	"	"	"	$CH_3NHCOCH(CH_3)O$	"	nD 1.5365(35.2°C)
110	"	"	"	$(CH_3)_2NCOCH(CH_3)O$	"	m.p. 148.9°C
111	"	"	"	HS	"	nD 1.5828(21.8°C)

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112	CH_3	CHF_2O	Cl	CH_3S	Cl	m.p. 95.7°C
113	"	"	"	C_2H_5S	"	m.p. 110.5°C
114	"	"	"	i-C_3H_7S	"	m.p. 83.8°C
115	"	"	"	n-C_4H_9S	"	m.p. 155.4°C
116	"	"	"	CHF_2S	"	m.p. 123.4°C
117	"	"	"	CH_2=$CHCH_2S$	"	m.p. 52.0 - 55.0°C
118	"	"	"	CH_2=$CHCH_2SO$	"	m.p. 91.5°C
119	"	"	"	$CH{\equiv}CCH_2S$	"	m.p. 127 - 129°C
120	"	"	"		"	nD 1.5972(20.2°C)
121	"	"	"	$(C_2H_5O)_2P(O)CH_2S$	"	nD 1.5245(25.3°C)
122	"	"	"	CH_3OOCS	"	nD 1.5656(17.2°C)
123	"	"	"	CH_3OOCCH_2S	"	m.p. 102.0°C
124	"	"	"	$C_2H_5OOCCH_2S$	"	m.p. 171.5°C
125	"	"	"	CH_2=$CHCH_2OOCCH_2S$	"	m.p. 102.4°C

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126	CH_3	CHF_2O	Cl	$HOOCCH(CH_3)S$	Cl	m.p. 188.9°C
127	"	"	"	$\overset{\oplus}{Na}\overset{\ominus}{O}OCCH(CH_3)S$	"	m.p. 205.4°C
128	"	"	"	$i\text{-}C_3H_7\overset{\oplus}{N}H_3\overset{\ominus}{O}OCCH(CH_3)S$	"	Viscous substance
129	"	"	"	$CH_3OOCCH(CH_3)S$	"	nD 1.5654(19.8°C)
130	"	"	"	$C_2H_5OOCCH(CH_3)S$	"	nD 1.5565(28.0°C)
131	"	"	"	$C_2H_5OOCCH(CH_3)S(O)$	"	nD 1.5582(20.0°C)
132	"	"	"	$C_2H_5OOCCH(CH_3)SO_2$	"	m.p. 105.8°C
133	"	"	"	$i\text{-}C_3H_7OOCCH(CH_3)S$	"	nD 1.5417(22.1°C)
134	"	"	"	$n\text{-}C_4H_9OOCCH(CH_3)S$	"	nD 1.5458(20.2°C)
135	"	"	"	$(CH_3)_2CH(CH_2)_2OOCCH(CH_3)S$	"	nD 1.5437(22.2°C)
136	"	"	"	$CH_2=CHCH_2OOCCH(CH_3)S$	"	nD 1.5600(20.4°C)
137	"	"	"	$CH\equiv CCH_2OOCCH(CH_3)S$	"	nD 1.5602(22.2°C)
138	"	"	"	$CH_3O(CH_2)_2O(CH_2)_2OOCCH(CH_3)S$	"	nD 1.5431(23.5°C)
139	"	"	"	$(C_2H_5O)_2P(O)CH_2OOCCH(CH_3)S$	"	nD 1.5386(21.2°C)

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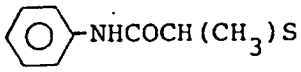
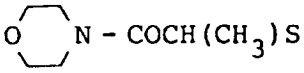
140	CH_3	CHF_2O	Cl	$CH_3NHCOCH(CH_3)S$	Cl	m.p. 99.9°C
141	"	"	"	$(CH_3)_2NCOCH(CH_3)S$	"	nD 1.5672(19.9°C)
142	"	"	"	$(C_2H_5)_2NCOCH(CH_3)S$	"	nD 1.5532(19.8°C)
143	"	"	"	$i\text{-}C_3H_7NHCOCH(CH_3)S$	"	m.p. 99.9°C
144	"	"	"		"	m.p. 132.3°C
145	"	"	"		"	nD 1.5457(19.9°C)
146	"	"	"	NO_2	"	m.p. 99.1°C
147	"	"	"	NH_2	"	nD 1.5743(20.8°C)
148	"	"	"	CH_3SO_2NH	"	m.p. 142.9°C
149	"	"	"	CH_3NHSO_2NH	"	m.p. 133.0°C
150	"	"	"	CH_3NH	"	nD 1.4952(25.2°C)
151	"	"	"	$(CH_3)_2N$	"	nD 1.5561(24.9°C)
152	"	"	"	$CH_2=CHCH_2NH$	"	nD 1.5719(25.3°C)
153	"	"	"	$CH\equiv CCH_2NH$	"	m.p. 76.3°C

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154	CH_3	CHF_2O	Cl	CH_3OOCNH	m.p. 162.3°C
155	"	"	"	$Cl^{\ominus}\ H_3N^{\oplus}$	m.p. 137.1°C
156	"	"	"		m.p. 244.1°C
157	"	"	"	CH_3OOCCH_2NH	m.p. 96.2°C
158	"	"	"	$HOOCCH(CH_3)NH$	m.p. 192.3°C
159	"	"	"	$Na^{\oplus}\ ^{\ominus}OOCCH(CH_3)NH$	m.p. 248.0 (decomp.)
160	"	"	"	$i\text{-}C_3H_7NH_3^{\oplus}\ ^{\ominus}OOCCH(CH_3)NH$	Viscous substance
161	"	"	"	$C_2H_5OOCCH(CH_3)NH$	nD 1.5371(23.4°C)
162	"	"	"	$CH_3NHOCCH(CH_3)NH$	nD 1.5500(25.6°C)
163	"	"	"	$(CH_3)_2NOCCH(CH_3)NH$	nD 1.5510(25.2°C)
164	"	"	"	CH_3NHSO_2	nD 1.5461(17.9°C)
165	"	"	"	$(CH_3)_2NSO_2$	m.p. 135.2°C
166	"	"	"	$(C_2H_5)_2NCOS$	nD 1.5623(17.7°C)

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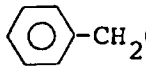
167	CH_3	CHF_2S	Cl	CH_3O	Cl	nD	1.5767(21.9°C)
168	"	"	"	C_2H_5O	"	nD	1.5810(23.8°C)
169	"	"	"	$i\text{-}C_3H_7O$	"	nD	1.5536(24.2°C)
170	"	"	"	$n\text{-}C_6H_{13}O$	"	nD	1.5460(24.2°C)
171	"	"	"	$CH_2{=}CHCH_2O$	"	nD	1.5791(19.8°C)
172	"	"	"	$CH{\equiv}CCH_2O$	"	nD	1.5720(20.0°C)
173	"	"	"		"	nD	1.6226(24.3°C)
174	"	"	"	CH_3OOCCH_2O	"	nD	1.5629(23.1°C)
175	"	"	"	$C_2H_5OOCCH(CH_3)O$	"	nD	1.5569(23.6°C)
176	"	"	"	HS	"	nD	1.6218(23.5°C)
177	"	"	"	CH_3S	"	nD	1.6069(21.7°C)
178	"	"	"	C_2H_5S	"	nD	1.6047(24.1°C)
179	"	"	"	$i\text{-}C_3H_7S$	"	nD	1.5893(23.6°C)
180	"	"	"	$n\text{-}C_5H_{11}S$	"	nD	1.5881(25.0°C)

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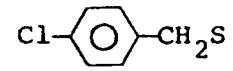
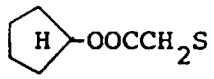
181	CH_3	CHF_2S	Cl	CHF_2S	Cl	nD 1.5869(24.2°C)
182	"	"	"	$CH_2=CHCH_2S$	"	nD 1.6173(21.3°C)
183	"	"	"	$CH\equiv CCH_2S$	"	nD 1.6261(23.4°C)
184	"	"	"	$C_2H_5OCH_2CH_2S$	"	nD 1.5816(22.3°C)
185	"	"	"	CH_3SCH_2S	"	nD 1.6222(24.2°C)
186	"	"	"	$NCCH_2S$	"	m.p. 95.9°C
187	"	"	"		"	nD 1.6273(24.3°C)
188	"	"	"	C_2H_5OOCS	"	nD 1.5720(21.8°C)
189	"	"	"	CH_3OOCCH_2S	"	m.p. 112.1°C
190	"	"	"	$C_2H_5OOCCH_2S$	"	m.p. 97.8°C
191	"	"	"		"	m.p. 104.4°C
192	"	"	"	$CH_3NHOCCH_2S$	"	m.p. 138.6°C
193	"	"	"	$(CH_3)_2NOCCH_2S$	"	m.p. 138.3°C

- cont'd -

194	CH_3	CHF_2S	Cl	$HOOCCH(CH_3)S$	Cl	m.p. 149.5°C
195	"	"	"	$NaOOCCH(CH_3)S$	"	m.p. 220.8°C
196	"	"	"	$i\text{-}C_3H_7NH_3\ OOCCH(CH_3)S$	"	Viscous substance
197	"	"	"	$C_2H_5OOCCH(CH_3)S$	"	nD 1.5788(23.4°C)
198	"	"	"	$n\text{-}C_3H_7OOCCH(CH_3)S$	"	nD 1.5643(19.4°C)
199	"	"	"	$i\text{-}C_3H_7OOCCH_2S$	"	nD 1.5695(19.4°C)
200	"	"	"	$(CH_3)_2CHCH_2OOCCH_2S$	"	nD 1.5812(21.2°C)
201	"	"	"	$CH_2=CHCH_2OOCCH(CH_3)S$	"	nD 1.5800(19.4°C)
202	"	"	"	$C_2H_5OOC(CH_2)_3S$	"	m.p. 110.7°C
203	"	"	"	$CH_3NHOCCH(CH_3)S$	"	m.p. 140.8°C
204	"	"	"	$(CH_3)_2NOCCH(CH_3)S$	"	nD 1.5791(21.0°C)
205	"	"	"	NO_2	"	m.p. 109.6°C
206	"	"	"	NH_2	"	nD 1.5837(20.3°C)
207	"	"	"	$(CH_3)_2N$	"	nD 1.5838(21.6°C)

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208	CH_3	CHF_2S	Cl	$CH_2=CHCH_2NH$	Cl	nD	1.6060(21.2°C)
209	"	"	"	$(CH_2=CHCH_2)_2N$	"	nD	1.5884(24.0°C)
210	"	"	"	$CH\equiv CCH_2NH$	"	nD	1.6052(23.1°C)
211	"	"	"	CH_3SO_2NH	"	m.p.	165.7°C
212	"	"	"	CH_3NHSO_2NH	"	nD	1.5812(20.4°C)
213	"	"	"	CH_3OOCNH	"	m.p.	144.5°C
214	"	"	"	$(C_2H_5)_2NCOS$	"	nD	1.5753(21.5°C)
215	"	CH_3S	"	$C_2H_5OOCCH(CH_3)O$	"	nD	1.5584(25.7°C) Dimethyl sulfate quaternary ammonium salt
216	"	CHF_2O	"	$(CH_3)_3SiCH_2S$	"	nD	1.5568(22.8°C)
217	"	CH_3SO	"	5-NO_2	F	m.p.	151.0°C
218	"	CH_3S	"	5-NH_2	"	nD	1.6260(27.3°C)
219	"	"	"	5-SH	"	Viscous substance	
220	"	"	"	5-$CH\equiv CCH_2O$	"	nD	1.5980(25.2°C)
221	"	"	"	5-$CH\equiv CCH_2S$	"	m.p.	93 - 96°C

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222	CH_3	CH_3S	Cl	5-$C_2H_5OOCCH(CH_3)S$	F	nD	1.5880(26.4°C)
223	"	SCH_3	"	CH_3O-$COCH_2O$	Cl	mp.	126.2°C
224	"	"	"	C_2H_5O-$COCH_2O$	"	mp.	106.5°C
225	"	$OCHF_2$	H	CH_2=$CHCH_2O$	"	mp.	56.2°C
226	"	"	"	CH≡CCH_2O	"	mp.	100.0°C
227	"	"	Cl	OHC	"	mp.	138.2°C
228	"	"	"	CH_2=$C(CH_3)CH_2O$	"	nD	1.5387(24.3°C)
229	"	"	"	$(CH_3)_3SiCH_2O$	"	nD	1.5271(23.9°C)
230	"	"	"	HO-$COCH_2O$	"	mp.	162.0°C
231	"	"	"	$Na^{\oplus}\ ^{\ominus}O$-$COCH_2O$	"	mp.	238.0°C
232	"	"	"	$K^{\oplus}\ ^{\ominus}O$-$COCH_2O$	"	mp.	194.0°C
233	"	"	"	i-$C_3H_7NH_3^{\oplus}\ ^{\ominus}O$-$COCH_2O$	"	mp.	131.0°C
234	"	"	"	C_2H_5O-$COCH_2O$	"	mp.	102.3°C
235	"	"	"	t-C_4H_9O-$COCH_2O$	"	mp.	92.2°C

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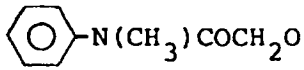
236	CH_3	$OCHF_2$	Cl	$CH_2=CHC(CH_3)_2O-COCH_2O$	Cl	mp.	92.8°C
237	"	"	"	$CH_3C\equiv CCH_2O-COCH_2O$	"	mp.	104.3°C
238	"	"	"	$CH_3S-COCH_2O$	"	mp.	117.5°C
239	"	"	"	$C_2H_5S-COCH_2O$	"	mp.	86.0°C
240	"	"	"	$i-C_3H_7S-COCH_2O$	"	mp.	98.0°C
241	"	"	"	$CH_3NHCOCH_2O$	"	mp.	149.7°C
242	"	"	"	$(CH_3)_2NCOCH_2O$	"	mp.	153.3°C
243	"	"	"	$n-C_4H_9NHCOCH_2O$	"	mp.	106.9°C
244	"	"	"	$H_2C=CHCH_2NHCOCH_2O$	"	mp.	127.7°C
245	"	"	"	$(H_2C=CHCH_2)_2NCOCH_2O$	"	mp.	80.0°C
246	"	"	"	 -$NHCOCH_2O$	"	mp.	205.6°C
247	"	"	"	 -$N(CH_3)COCH_2O$	"	nD	1.5570(18.7°C)
248	"	"	"	$CH_3SO_2NHCOCH_2O$	"	mp.	124.5°C
249	"	"	"	$(CH_3)_3SiCH_2O-COCH_2O$	"	nD	1.5223(29.5°C)

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250	CH_3	$OCHF_2$	Cl	$CH_3OC_2H_4OC_2H_4O$-$COCH(CH_3)O$	Cl	nD	1.5142(23.9°C)
251	"	"	"	C_2H_5O-$COCH(C_2H_5)O$	"	nD	1.5286(26.5°C)
252	"	"	"	C_2H_5O-$CO(CH_2)_3O$	"	nD	1.5238(17.3°C)
253	"	"	"	CH_3O-$COCH(i\text{-}C_3H_7)O$	"	nD	1.5273(24.3°C)
254	"	"	"	$CH_3SO_2NHCOCH(CH_3)O$	"	mp.	51.6°C
255	"	"	"	OHC	F	mp.	107.3°C
256	"	"	"	CH_2=$CHCH_2O$	"	mp.	63.7 - 64.1°C
257	"	"	"	CH_2=$C(CH_3)CH_2O$	"	nD	1.5376(17.3°C)
258	"	"	"	CH≡CCH_2O	"	mp.	98.0 - 98.1°C
259	"	"	"	CH_3C≡CCH_2O	"	mp.	108.5°C
260	"	"	"	HO-$COCH_2O$	"	mp.	143.7°C
261	"	"	"	CH_3O-$COCH_2O$	"	mp.	122.8 - 123.1°C
262	"	"	"	C_2H_5O-$COCH_2O$	"	mp.	127.6°C
263	"	"	"	$n\text{-}C_3H_7O$-$COCH_2O$	"	mp.	97.6 - 97.8°C
264	"	"	"	$i\text{-}C_3H_7O$-$COCH_2O$	"	mp.	120.3 - 120.5°C

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265	CH_3	$OCHF_2$	Cl	$n\text{-}C_4H_9O\text{-}COCH_2O$	F	mp. 91.4 - 91.6°C
266	"	"	"	$CH_2{=}CHCH_2O\text{-}COCH_2O$	"	mp. 89.2 - 89.4°C
267	"	"	"	$CH{\equiv}CCH_2O\text{-}COCH_2O$	"	mp. 99.0°C
268	"	"	"	$C_2H_5S\text{-}COCH_2O$	"	mp. 69.3°C
269	"	"	"	$i\text{-}C_3H_7S\text{-}COCH_2O$	"	mp. 83.8°C
270	"	"	"	$HO\text{-}COCH(CH_3)O$	"	mp. 94.5°C
271	"	"	"	$CH_3O\text{-}COCH(CH_3)O$	"	mp. 95.6°C
272	"	"	"	$C_2H_5O\text{-}COCH(CH_3)O$	"	mp. 67.0 - 67.2°C
273	"	"	"	$CH_2{=}CHCH_2O\text{-}COCH(CH_3)O$	"	Viscous substance
274	"	"	"	$CH{\equiv}CCH_2O\text{-}COCH(CH_3)O$	"	nD 1.5266(25.8°C)
275	"	"	"	$C_2H_5O\text{-}COCH(C_2H_5)O$	"	nD 1.5105(22.6°C)
276	"	"	"	$CH_2{=}C(CH_3)CH_2S$	Cl	nD 1.5775(21.7°C)
277	"	"	"	$CH_2{=}C(Cl)CH_2S$	"	nD 1.5865(21.5°C)
278	"	"	"	$ClCH{=}CHCH_2S$	"	nD 1.5867(21.2°C)
279	"	"	"	$ClCH{=}C(Cl)CH_2S$	"	nD 1.5926(22.8°C)
280	"	"	"	$NCCH_2S$	"	mp. 122.3°C

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281	CH_3	$OCHF_2$	Cl	$\oplus\ominus$ Na $O-COCH_2S$	mp.	232.0°C
282	"	"	"	$(i-C_3H_7)NH_3^{\oplus}\ O-COCH_2S^{\ominus}$	mp.	113.0°C
283	"	"	"	$HO-COCH_2S$	mp.	164.2°C
284	"	"	"	$i-C_3H_7O-COCH_2S$	mp.	99.1°C
285	"	"	"	$n-C_4H_9O-COCH_2S$	mp.	62.0°C
286	"	"	"	$C_{12}H_{25}O-COCH_2S$	nD	1.5010(27.2°C)
287	"	"	"	$CH_3S-COCH_2S$	mp.	98.3°C
288	"	"	"	$C_2H_5S-COCH_2S$	mp.	73.1°C
289	"	"	"	$i-C_3H_7S-COCH_2S$	mp.	97.0°C
290	"	"	"	$CH\equiv CCH_2O-COCH_2S$	mp.	85.9°C
291	"	"	"	$C_2H_5OC_2H_4OC_2H_4O-COCH_2S$	mp.	76.5°C
292	"	"	"		mp.	87.8°C
293	"	"	"		mp.	82.8°C

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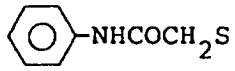
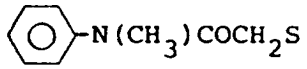
294	CH_3	$OCHF_2$	Cl	$(CH_3)_3SiCH_2O\text{-}COCH_2S$	Cl	nD 1.5387(29.1°C)
295	"	"	"	$CH_3NHCOCH_2S$	"	mp. 177.8°C
296	"	"	"	$(CH_3)_2NCOCH_2S$	"	mp. 121.6°C
297	"	"	"	$H_2C{=}CHCH_2NHCOCH_2S$	"	mp. 146.6°C
298	"	"	"	$(H_2C{=}CHCH_2)_2NCOCH_2S$	"	mp. 71.8°C
299	"	"	"	$HC{\equiv}CCH_2NHCOCH_2S$	"	mp. 172.0°C
300	"	"	"		"	mp. 178.1°C
301	"	"	"		"	nD 1.5661(21.5°C)
302	"	"	"		"	mp. 120.4°C
303	"	"	"		"	mp. 126.5°C
304	"	"	"	$i\text{-}C_4H_9O\text{-}COCH(CH_3)S$	"	Viscous substance
305	"	"	"	$t\text{-}C_4H_9CH_2O\text{-}COCH(CH_3)S$	"	nD 1.5462(13.9°C)

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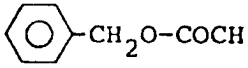
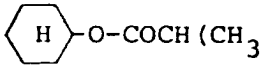
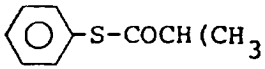
306	CH_3	$OCHF_2$	Cl	$n\text{-}C_6H_{13}O\text{-}COCH(CH_3)S$	Cl	nD 1.5210(23.2°C)
307	"	"	"		"	Viscous substance
308	"	"	"	$C_2H_5S\text{-}COCH(CH_3)S$	"	nD 1.5790(24.0°C)
309	"	"	"	$i\text{-}C_3H_7S\text{-}COCH(CH_3)S$	"	nD 1.5669(24.2°C)
310	"	"	"	$NCCH(CH_3)O\text{-}COCH(CH_3)S$	"	nD 1.5501(23.1°C)
311	"	"	"	$CH_3SC_2H_4O\text{-}COCH(CH_3)S$	"	nD 1.5609(23.1°C)
312	"	"	"		"	Viscous substance
313	"	"	"		"	nD 1.5956(14.3°C)
314	"	"	"	$(CH_3)_3SiCH_2O\text{-}COCH(CH_3)S$	"	nD 1.5429(26.8°C)
315	"	"	"	$CH_2{=}CHCH_2NHCOCH(CH_3)S$	"	nD 1.5619(22.6°C)
316	"	"	"	$(H_2C{=}CHCH_2)_2NCOCH(CH_3)S$	"	nD 1.5603(22.9°C)
317	"	"	"	$HC{\equiv}CCH_2NHCOCH(CH_3)S$	"	nD 1.5496(23.1°C)
318	"	"	"	$CH_3CO\text{-}O\text{-}(CH_2)_2S$	"	nD 1.5603(26.9°C)
319	"	"	"	$C_2H_5O\text{-}COC(CH_3)_2S$	"	mp. 96.4°C

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320	CH_3	$OCHF_2$	Cl	CH_3O-COCH(i-C_3H_7)S	Cl	nD	1.5572(23.4°C)
321	"	"	"	C_2H_5O-COCH(CH_3)N(CH_3)	"	nD	1.5365(22.8°C)
322	"	"	"	HO-CO	"	mp.	216.7°C
323	"	"	"	CH_3O-CO	"	mp.	63.9°C
324	"	"	"	C_2H_5O-CO	"	nD	1.5446(26.8°C)
325	"	"	"	CH_2=$CHCH_2O$-CO	"	nD	1.5317(26.8°C)
326	"	"	"	CH_3O-COCH(CH_3)O-CO	"	nD	1.5370(25.7°C)
327	"	"	"	C_2H_5O-COCH(CH_3)O-CO	"	nD	1.5672(26.0°C)
328	"	"	"	CH≡CCH_2O-CO	"	mp.	78.5°C
329	"	"	Br	$BrCH_2CH(Br)CH_2O$	"	nD	1.5862(22.3°C)
330	"	"	"	BrC≡CCH_2O	"	nD	1.5603(22.9°C)
331	"	"	"	CH_3O-$COCH_2O$	"	mp.	133.8°C
332	"	"	"	C_2H_5O-COCH(CH_3)O	"	nD	1.5396(20.8°C)
333	"	$SCHF_2$	Cl	NCCH_2O	"	nD	1.5571(26.1°C)
334	"	"	"	CH_2=C(CH_3)CH_2O	"	nD	1.5767(26.3°C)

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335	CH_3	$SCHF_2$	Cl	$ClCH=CHCH_2O$	Cl	nD	1.5930(25.8°C)
336	"	"	"	CH_3OCH_2O	"	nD	1.5765(23.8°C)
337	"	"	"	$C_2H_5OC_2H_4O$	"	nD	1.5713(23.4°C)
338	"	"	"	CH_3OCH_2S	"	mp.	82.6°C
339	"	"	"	$ClCH=CHCH_2S$	"	nD	1.6194(26.3°C)
340	"	"	"	$CH_2=C(Cl)CH_2S$	"	nD	1.6137(26.2°C)
341	"	$OCHF_2$	"	HS	F	mp.	72.6°C
342	"	"	"	CH_3S	"	mp.	84.5°C
343	"	"	"	F_2HCS	"	nD	1.5377(24.0°C)
344	"	"	"	$CH_2=CHCH_2S$	"	nD	1.5670(17.9°C)
345	"	"	"	$CH_2=C(CH_3)CH_2S$	"	nD	1.5637(19.1°C)
346	"	"	"	$CH\equiv CCH_2S$	"	mp.	82.8°C
347	"	"	"	$C_2H_5OC_2H_4S$	"	nD	1.5523(24.0°C)
348	"	"	"	$HO-COCH_2S$	"	mp.	132.2°C
349	"	"	"	$CH_3O-COCH_2S$	"	nD	1.5579(24.0°C)

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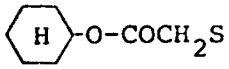
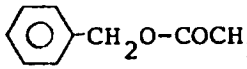
350	CH_3	$OCHF_2$	Cl	$C_2H_5O-COCH_2S$	F	nD	1.5342(17.9°C)
351	"	"	"	$C_2H_5O-COCH_2S(O)$	"	nD	1.5420(25.0°C)
352	"	"	"	$n-C_4H_9O-COCH_2S$	"	nD	1.5391(24.1°C)
353	"	"	"	$CH_2=CHCH_2O-COCH_2S$	"	nD	1.5562(24.1°C)
354	"	"	"	$CH_3SCH_2O-COCH_2S$	"	mp.	57.9°C
355	"	"	"		"	mp.	58.9°C
356	"	"	"		"	nD	1.5794(17.9°C)
357	"	"	"	$CH_3NHCOCH_2S$	"	mp.	147.0°C
358	"	"	"	$(CH_3)_2NCOCH_2S$	"	mp.	141.2°C
359	"	"	"	$HO-COCH(CH_3)S$	"	nD	1.5463(19.1°C)
360	"	"	"	$CH_3O-COCH(CH_3)S$	"	nD	1.5494(25.0°C)
361	"	"	"	$C_2H_5O-COCH(CH_3)S$	"	nD	1.5328(18.0°C)
362	"	"	"	$n-C_4H_9O-COCH(CH_3)S$	"	nD	1.5343(25.1°C)

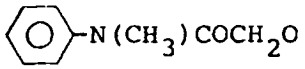
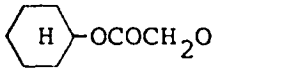
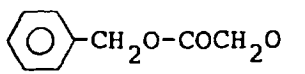
- cont'd -

363	CH_3	$OCHF_2$	Cl	$CH_2=CHCH_2O-COCH(CH_3)S$	F	nD 1.5471(25.0°C)
364	"	"	"	$CH\equiv CCH_2O-COCH(CH_3)S$	"	Viscous substance
365	"	"	"	$CH_3OC_2H_4OC_2H_4O-COCH(CH_3)S$	"	nD 1.5355(18.4°C)
366	"	"	"	CH_3SO_2NH	"	mp. 146.9°C
367	"	"	"	CH_3NHSO_2NH	"	mp. 131.2°C
368	"	"	"	$CH_2=CHCH_2NH$	"	nD 1.5424(23.6°C)
369	"	"	"	$CH\equiv CCH_2NH$	"	nD 1.5510(27.1°C)
370	"	"	"	$C_2H_5O-COCH_2NH$	"	mp. 111.5°C
371	"	"	"	$C_2H_5O-COCH(CH_3)NH$	"	nD 1.5264(26.6°C)
372	"	"	"	CH_3O-CO	"	nD 1.5430(17.0°C)
373	"	"	"	C_2H_5O-CO	"	nD 1.5320(21.0°C)
374	"	"	"	$CH_2=CHCH_2O-CO$	"	nD 1.5218(20.5°C)
375	"	"	"	$CH\equiv CCH_2O-CO$	"	nD 1.5820(24.5°C)
376	"	"	"	$CH_3O-COCH(CH_3)O-CO$	"	nD 1.5314(23.0°C)
377	"	"	"	$C_2H_5O-COCH(CH_3)O-CO$	"	nD 1.5212(14.1°C)

- cont'd -

378	CH_3	$SCHF_2$	Cl		Cl	mp. 102.9°C
379	"	"	"		"	mp. 104.4°C
380	"	"	"	$HO-COCH_2O$	"	mp. 157.7°C
381	"	"	"	$\oplus\ \ominus$ $Na\ O-COCH_2O$	"	mp. 135.2°C
382	"	"	"	$C_2H_5O-COCH_2O$	"	nD 1.5736(24.0°C)
383	"	"	"	$i-C_3H_7O-COCH_2O$	"	mp. 113.3°C
384	"	"	"	$CF_3CH_2O-COCH_2O$	"	mp. 117.5°C
385	"	"	"	$CH_2=CHCH_2O-COCH_2O$	"	mp. 77.2°C
386	"	"	"		"	nD 1.5619(28.3°C)
387	"	"	"	$CH_3NHCOCH_2O$	"	mp. 116.8°C
388	"	"	"	$CH_3O-COCH(CH_3)O$	"	nD 1.5620(23.4°C)
389	"	"	"	$i-C_3H_7O-COCH(CH_3)O$	"	nD 1.5496(24.5°C)

- cont'd -

390	CH_3	$SCHF_2$	Cl	$CH_2=CHCH_2O-COCH(CH_3)O$	Cl	nD	1.5643(23.5°C)
391	"	"	"	$CH_3NHCOCH(CH_3)O$	"	nD	1.5641(23.5°C)
392	"	"	"	$CH_3O-COCH(n-C_4H_9)O$	"	nD	1.5488(28.1°C)
393	"	"	"	$NCCH_2O-COCH_2O$	"	nD	1.5694(28.0°C)
394	"	"	"	$CH_3O-COCH(CH_3)S$	"	nD	1.5891(26.4°C)
395	"	"	"	$NCCH_2O-COCH(CH_3)S$	"	nD	1.5801(26.5°C)
396	"	"	"	$C_2H_5O-CO(CH_2)_3S$	"	mp.	107.0°C
397	"	"	"		"	mp.	136.9°C
398	"	"	"	$HO-COCH_2NH$	"	mp.	204.0°C (decomp.)
399	"	"	"	$C_2H_5O-COCH_2NH$	"	mp.	110.0°C
400	"	"	"	$C_2H_5O-COCH(CH_3)NH$	"	nD	1.5730(23.9°C)
401	"	$OCHF_2$	"		"	mp.	91.8°C
402	"	"	"		"	mp.	139.1°C

- cont'd -

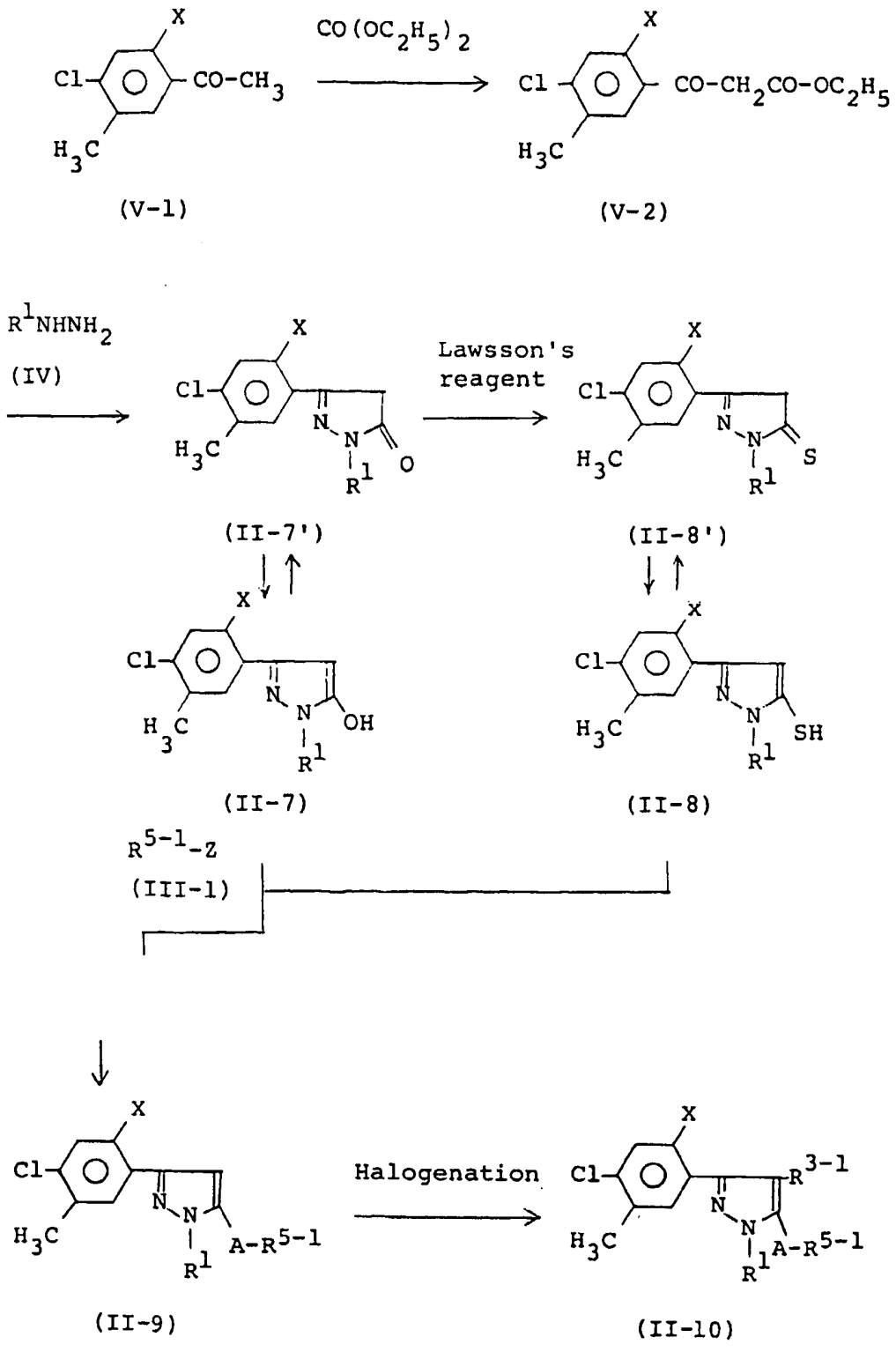
403	CH_3	$OCHF_2$	Cl	HO-CO	F	Solid substance	
404	"	"	"	NO_2	"	"	
405	"	"	"	NH_2	"	Viscous substance	
406	"	"	"	HO- $\oplus\ominus$ Na O-CO	"	Solid substance	
407	"	"	"	"	Cl	mp. 246.4°C	
408	"	"	"	CH_3O-CO-OCH_2S-CO	"	nD 1.5727(24.1°C)	
409	"	"	"	$(CH_3)_2NCO\ \oplus\ominus$	F	Viscous substance	
410	"	"	"	$i\text{-}C_3H_7NH_4CO$	"	nD 1.5269(23.5°C)	

Table 2 shows NMR data on viscous or crystalline substances shown in Table 1.

Table 2

Comp'd No.	NMR (δ value, $CDCl_3$/TMS, ppm
19	2.38 (3H, s), 3.50 (2H, br), 3.96 (3H, s), 4.93 - 5.33 (2H, m), 5.46 - 6.23 (1H, m), 7.25 (1H, s), 7.43 (1H, s).
64	1.23 (3H, t), 1.51 (3H, d), 2.40 (3H, s), 3.97 (3H, s), 4.18 (2H, q), 3.80 - 4.16 (1H, m), 4.90 (1H, d), 6.64 (1H, s), 7.40 (1H, s).
104	1.13 (6H, d), 1.58 (3H, d), 2.83 - 3.45 (1H, m), 3.80 (3H, s), 4.59 (1H, q), 6.75 (1H, t), 6.92 (1H, s), 7.51 (1H, s), 7.75 - 8.35 (3H, br).
128	1.18 (6H, d), 1.49 (3H, d), 2.92 - 3.58 (1H, m), 3.81 (3H, s), 4.06 (1H, q), 6.68 (1H, t), 7.43 (2H, s), 7.62 - 8.32 (3H, br).
160	1.12 (6H, d), 1.48 (3H, d), 2.60 - 3.30 (1H, m), 3.73 (3H, s), 3.91 (1H, q), 6.55 (1H, s), 6.68 (1H, s), 7.33 (1H, s), 7.98 (3H, s).
196	1.20 (6H, d), 1.48 (3H, d), 3.28 (1H, m), 3.90 (1H, m), 4.00 (3H, s), 6.74 (1H, t, J=56 Hz), 7.45 (2H, s), 8.00 (3H, s).
219	2.28 (3H, t), 3.91 (1H), 3.99 (3H, s), 7.21 (1H, d), 7.50 (1H, d).
273	1.70 (3H, d), 3.80 (3H, s), 4.5 - 4.9 (2H), 5.0 - 5.5 (2H), 5.5 - 5.2 (1H), 6.70 (1H, t, J=73 Hz), 7.0 - 7.4 (2H).
304	0.82 (6H, d), 1.56 (3H, d), 3.80 (2H, d), 3.73 - 4.23 (2H, m), 6.70 (1H, t), 7.20 (1H, s), 7.23 (1H,s).
307	1.53 (3H, d), 3.75 (3H, s), 4.06 (1H, q), 5.10 (2H, s), 6.61 (1H, t), 7.18 (5H, s), 7.41 (1H, s).
312	1.53 (3H, d), 0.68 - 2.11 (10H, m), 3.80 (3H, s), 4.10 (1H, q), 4.33 - 4.90 (1H, br), 6.70 (1H, t), 7.20 (1H, s), 7.23 (1H, s).
364	1.54 (3H, d), 2.42 (1H, t), 3.80 (3H, s), 3.98 (1H, q), 4.61 (2H, d), 6.71 (1H, t, J=72 Hz), 7.31 (1H, d, J=9 Hz), 7.80 (1H, d, J=7.6 Hz).
404 ($CDCl_3$+d_6· DMSO)	3.80 (3H, s), 6.70 (1H, t, J=72 Hz), 7.22 (1H, d, J=10 Hz), 8.12 (1H, d, J=8 Hz).
405	3.83 (3H, s), 6.67 (1H, t, J=72 Hz), 7.36 (1H, d, J=10 Hz), 8.20 (1H, d, J=8 Hz).
406	3.73 (3H, s), 3.85 (2H, s, br), 6.64 (1H, t, J=72 Hz), 6.82 (1H, d, J=8 Hz), 7.04 (1H, d, J=10 Hz).
407	3.80 (3H, s), 6.00 (1H, Br), 6.65 (1H, t, 72 Hz), 7.01 (1H, d, J=10 Hz), 7.01 (1H, d, J=8 Hz)
410	3.10 (6H, s), 3.80 (3H, s), 6.70 (1H, t, J=72 Hz), 7.23 (1H, d, J=10 Hz), 7.53 (1H, d, J=8 Hz)

Pyrazole derivatives of formula (II-2) that are starting materials for 3-(substituted phenyl)-pyrozole derivatives of formula (I) or salts thereof can be produced, for instance, according to the following reaction schemes. Pyrazole derivatives of formula (II-2), in connection with their raw materials, are classified into groups of formulae (II-7), (II-8), (II-9), and (II-10).



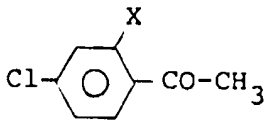
In the above equations, R^1, R^2, R^5, X, and Z are as defined above, R^{3-1} denotes halogen, and R^{5-1} denotes alkyl or haloalkyl.

As shown above, a pyrazole derivative of formula (II-8') can be prepared by reacting a compound of formula (V-1) with diethyl carbonate, reacting the resulting compound of formula (V-2) with a hydrazine of formula (IV), and reacting the resulting pyrazole derivative of formula (II-7') with Lawsson's reagent.

The pyrazole derivative of formula (II-10) can be produced from a pyrazole derivative of formula (II-7) that is the tautomer of pyrazole derivative of formula (II-7') or from a pyrazole derivative of formula (II-8) that is the tautomer of pyrazole derivative of formula (II-8') by reacting a halide of formula (III-1), followed by halogenating the resulting pyrazole derivative of formula (II-9).

Thus, the pyrazole derivatives of formula (II-2) can be produced that are raw materials used in the above production process①

Pyrazole derivatives of formula (II-5) that are raw materials in the process②A can be produced from compounds of the following formula (V-3) in the same manner as stated above.



(V-3)

Table III shows typical examples of the compounds of formula (II-2) which have been prepared according to the above process

Formula (II-2):

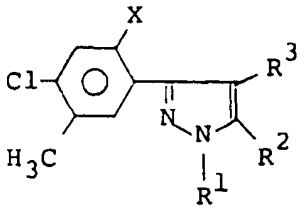


Table 3

Comp'd No.	R^1	R^2	R^3	X	Property
II.1	CH_3	OCH_3	Cl	F	nD 1.5633(26.5°C)
II.2	CH_3	$OCHF_2$	H	Cl	nD 1.5467(24.4°C)
II.3	CH_3	$OCHF_2$	H	F	Oily matter
II.4	CH_3	$OCHF_2$	Cl	Cl	nD 1.5511(24.5°C)
II.5	CH_3	$OCHF_2$	Cl	F	mp. 93.7°C
II.6	CH_3	$OCHF_2$	Br	Cl	nD 1.5627(24.8°C)
II.7	CH_3	$OCHF_2$	Br	F	mp. 126.4°C
II.8	i-C_3H_7	SCH_3	H	F	Oily matter
II.9	i-C_3H_7	SCH_3	Cl	F	nD 1.5830(15.1°C)
II.10	t-C_4H_9	$OCHF_2$	Cl	F	nD 1.5402(24.8°C)
II.11	t-C_4H_9	$OCHF_2$	Br	F	nD 1.4848(24.2°C)

Table 4 shows typical examples of the pyrazole derivatives of formula (II-5)

Formula (II-5):

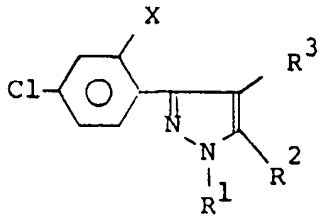


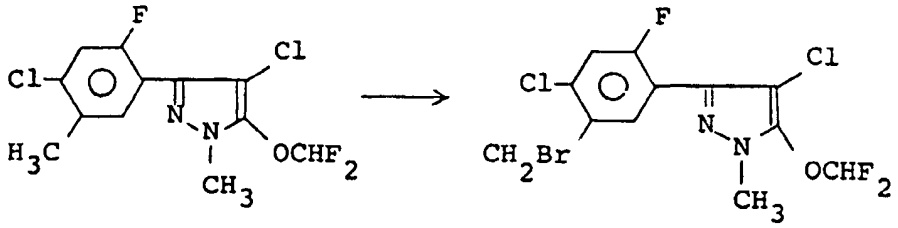
Table 4

Comp'd No.	R^1	R^2	R^3	X	Property
II.12	CH_3	OCH_3	H	Cl	mp. 79.4°C
II.13	CH_3	SCH_3	H	Cl	mp. 59.5 - 60.0°C
II.14	CH_3	SCH_3	H	F	mp. 40.8°C
II.15	CH_3	OCH_3	Cl	Cl	mp. 71 - 73°C
II.16	CH_3	SCH_3	Cl	Cl	mp. 62.5 - 63.5°C
II.17	CH_3	SCH_3	Cl	F	nD 1.6066(21.5°C)
II.18	CH_3	OCH_3	Br	Cl	mp. 83 - 85°C
II.19	CH_3	SCH_3	Br	Cl	mp. 70.0 - 75.5°C
II.20	CH_3	$OCHF_2$	H	Cl	mp. 39.2°C
II.21	CH_3	$SCHF_2$	H	Cl	nD 1.6001 (20.2°C)
II.22	CH_3	$OCHF_2$	H	F	mp. 41.2 - 41.5°C
II.23	CH_3	$OCHF_2$	Cl	Cl	mp. 27 - 28°C
II.24	CH_3	$SCHF_2$	Cl	Cl	nD 1.5900(20.2°C)
II.25	CH_3	$OCHF_2$	Cl	F	mp. 35.9 - 36.2°C)
II.26	CH_3	$OCHF_2$	Br	Cl	nD 1.5664(21.0°C)
II.27	CH_3	$SCHF_2$	Br	Cl	nD 1.5993(18.3°C)
II.28	CH_3	$OCHF_2$	Br	F	mp. 63.9°C
II.29	CH_3	$OCHF_2$	F	Cl	nD 1.5441(20.8°C)
II.30	CH_3	OCF_2CHF_2	H	Cl	nD 1.5237(20.5°C)
II.31	CH_3	OCF_2CHF_2	Cl	Cl	nD 1.5283(10.6°C)
II.32	CH_3	OCF_2CF_2Cl	Cl	Cl	mp. 68 - 70°C
II.33	CH_3	OCF_2CHFCF_3	H	Cl	mp. 68 - 70°C
II.34	C_2H_5	$OCHF_2$	Cl	Cl	nD 1.5422(27.3°C)

The present invention is illustrated with reference to typycal examples thereof.

Example 1 Preparation of 2-chloro-5-(4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazol-3-yl)-4-benzoic acid (compound No. 404)

1-1 Preparation of 3-(5-bromomethyl-4-chloro-2-fluorophenyl)-4-chloro-5-difluoromethoxy-1-methyl-IH-pyrazole

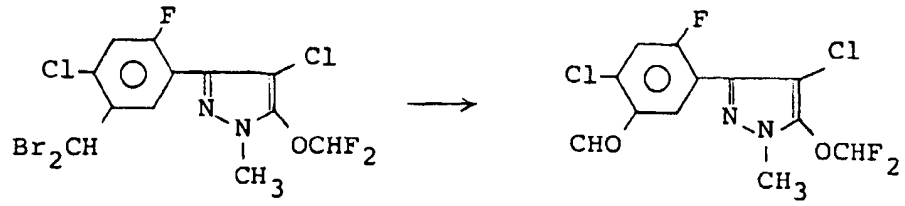


4-Chloro-3-(4-chloro-2-fluorc-5-methylphenyl)-5-difluoromethoxy-l-methyl-lH-pyrazole (6.9 g, 20 mmoles) was suspended in 100 ml carbon tetrachloride, and N-bromosuccinimide (3.97 g, 22 mmoles) together with a catalytic amount of benzoyl peroxide was added and reacted with the pyrazole derivative under reflux for 5 hours. Then the removal of insoluble matter from the product solution by filtration, concentration of the filtrate, and purification of the residue by column chromatography gave the title compound (5.64 g) as a viscous substance, yield 70%.

NMR ($CDCl_3$/TMS), δ (ppm):

3.83 (3H, s), 4.53 (2H, s), 6.67 (1H, t, J=72 Hz), 7.22 (1H, d, J=10 Hz), 7.60 (1H, d, J=8 Hz).

1-2 Preparation of 2-chloro-(4-chlor--5-difluoromethoxy-l-methyl-1H-pyrazol-3-yl)-4-fluorobenzaldehyde (compound No. 255)

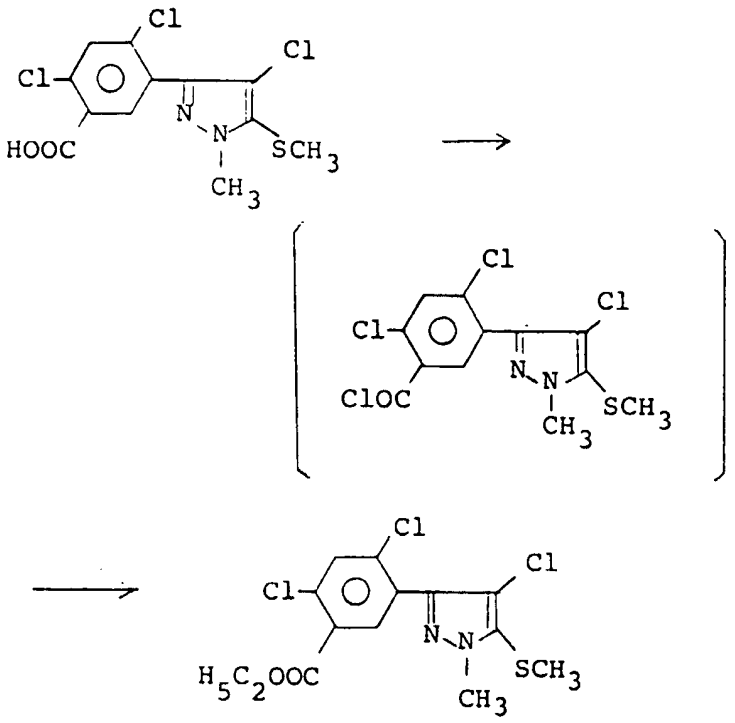


4-Chloro-3-(4-chloro-5-dibromomethyl-2-fluorophenyl)-5-difluromethoxy-1-methyl-1H-pyrazole (14.4 g, 30 mmoles) was suspended in 50 ml conc. hydrochloric acid and reacted under reflux for 5 hours. Then the reaction product solution, poured into ice-cold water, was extracted with ethyl acetate. The extract solution was washed successively with 5% aqueous $NaHCO_3$ and water, and dehydrated and concentrated. The residue was purified by column chromatography, giving the title compound (6.0 g), m.p. 107.3°C, yield 59%.

1-3 Preparation of 2-chloro-5-(4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazol-3-yl)-4-fluorobenzoic acid (compound No. 403)

A solution of potassium permanate (1.1 g, 7 mmoles) in 22 ml water was dropped slowly into a suspension of 2-chloro-5-(4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazol-3-yl)-4-fluorobenzaldehyde (1.7 g, 5 mmoles) in 18 ml water at 70 - 80°C to react. After dropping was finished, the reaction was further continued for 1 hour at 70 - 80°C. Then the reaction product solution was cooled to room temperature, and made alkaline with 10% aqueous NaOH. The by-product manganese dioxide was filtered off, and washed with hot water. The filtrate and the washing were mixed together, acidified with hydrochloric acid, and extracted with ether. The ether layer was washed with water, dehydrated, and evaporated to dryness, giving the title compound (1.0 g) in block form, yield 56%.

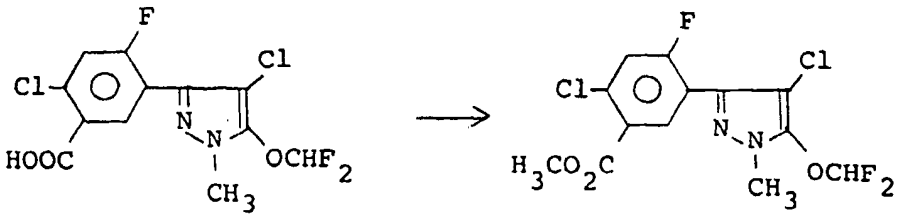
Example 2 Preparation of ethyl 5-(4-chloro-1-methyl-5-methylthio-1H-pyrazol-3-yl)-2,4-dichlorobenzoate (compound No. 50)



Ethyl 5-(4-chloro-1-methyl-5-methylthio-1H pyrazol-3-yl)-2,4-dichlorobenzoate (0.30 g, 0.85 mmole) and thionyl chloride (0.10 g, 0.85 mmole), mixed with 20 ml methylene chloride, were reacted together under reflux for 2 hours. Then the solvent was evaporated away under reduced pressure to give an acid chloride, which in turn was reacted with a large excess of ethanol under reflux for 1.5 hours. Then, romoval of the solvent by evaporation under reduced pressure and purification of the resulting residue by silica gel column chromatography gave the title compound (0.11 g) in paste form, yield 33.9%.

nD(20.1°C): 1.6029

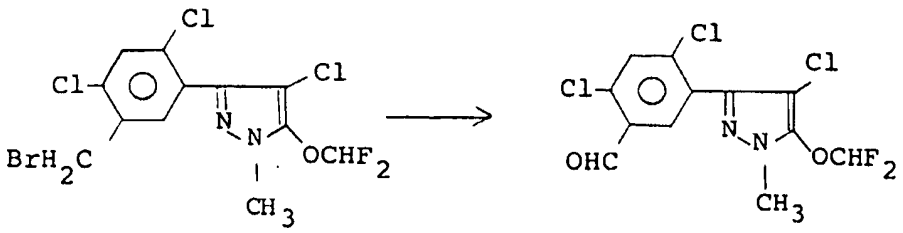
Example 3 Preparation of methyl 2-chloro-5-difluoromethoxy-1-methyl-1H-pyrazol-3-yl)-4-fluorobenzoate (compound No. 372)



A KOH powder (0.1 g, 1.8 mmoles) and methyl iodide (0.26 g, 1.8 mmols) were added to a solution of 2-chloro-5-(4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazol-3-yl)-4-fluorobenzoic acid in 20 ml acetone and reacted with this pyrazole derivative under reflux 3 hours. Then the extraction with ethylacetate, washing with water, dehydration, and concentration of the extract solution, and purification of the residue by column chromatography gave the title compound (0.15 g), yield 48%.

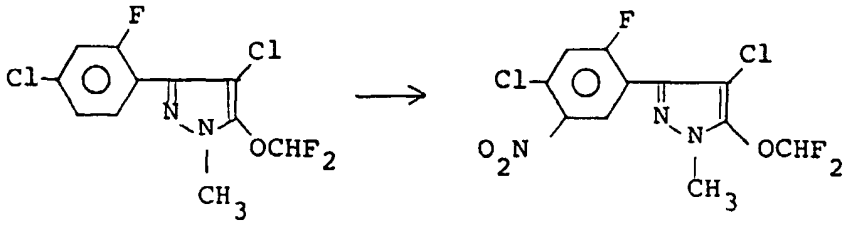
nD (17.0°C): 1.5430

Example 4 Preparation of 5-(4-chloro-5-difluoromethoxy-l-methyl-lH-pyrazol-3-yl)-2,4-dichlorobenzaldehyde (compound No. 227)



After addition of 100 ml water to a solution of 3-(5-bromomethyl-2,4-dichlorophenyl)-4-chloro-5-difluoromethoxy-l-methyl-1H-pyrazole (26.60 g, 63.3 mmoles) in 100 ml glacial acetic acid, hexamethylenetetramine (8.87 g, 63.3 mmoles) was added and reacted under reflux for 2 hours. Further, 40 ml conc. sulfuric acid was added and reacted under reflux for 13 hours. Then the reaction product mixture, poured into ice-cold water, was extracted with ethyl acetate. The extract solution was washed with 5% aqueous $NaHCO_3$ and then with water, dehydrated, and concentrated. Purification of the residue by column chromatography gave the title compound (12.66 g), m.p. 138.2°C, yield 56.3%.

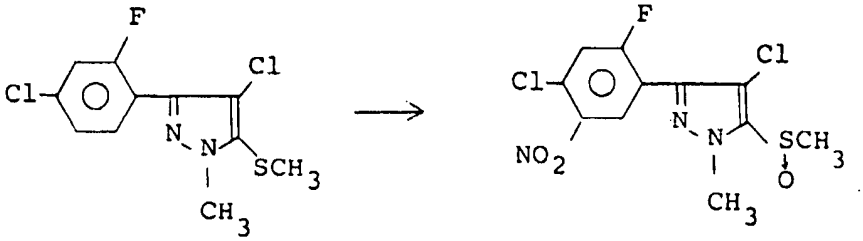
Example 5 Preparation of 4-chloro-3-(4-chloro-2-fluoro-5-nitrophenyl)-5-difluoromethoxy-1-methyl-1H-pyrazole (compound No. 404)



A mixture of 60% nitric acid (1.58 g, 0.015 mole) and conc. sulfuric acid (4.90 g, 0.05 mole) was added dropwise to a solution of 4-chloro-3-(4-chloro-2-fluorophenyl)-5-difluoromethoxy-1-methyl-1H-pyrazole (3.11 g, 0.01 mole) in 6 ml conc. sulfuric acid at temperatures of -10 to 10°C. Thereafter the reaction mixture was further stirred for 3 hours at room temperature. The reaction product mixture, poured into ice-cold water, was extracted with ethyl acetate. The ethyl acetate layer was washed successively with 5% aqueous $NaHCO_3$ and water, dehydrated, and evaporated to dryness, giving the title compound (3.0 g), yield 84%.

NMR, δ (ppm): 3.83 (3H, s), 6.67 (1H, t, J=72 Hz), 7.36 (1H, d, J=10 Hz), 8.20 (1H, d, J=8 Hz).

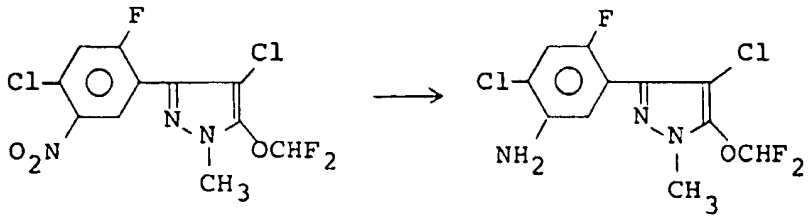
Example 6 Preparation of 4-chloro-3-(4-chloro-2-fluoro-5-nitrophenyl)-1-methyl-5-methylsulfinyl-1H-pyrazole (compound No. 217)



A mixture of conc. sulfuric acid (1.65 g) and conc. nitric acid (sp. gr. 1.38, 8.4 mmoles) was added dropwise to a

solution of 4-chloro-3-(4-chloro-2-fluorophenyl)-1-methyl-5-methylthio-1H-pyrazole (0.98 g, 3.4 mmoles) in 2 ml conc. sulfuric acid under cooling with ice. After the addition was finished, the reaction mixture was further stirred for 2 hours at room temperature, and allowed to stand overnight. Then the resulting mixture was poured into ice-cold water, and the formed crystals were filtered off, washed successively with water and ether, and dried, giving crystals (1.04 g) of the title compound, m.p. 151.0°C, yield 87.5%.

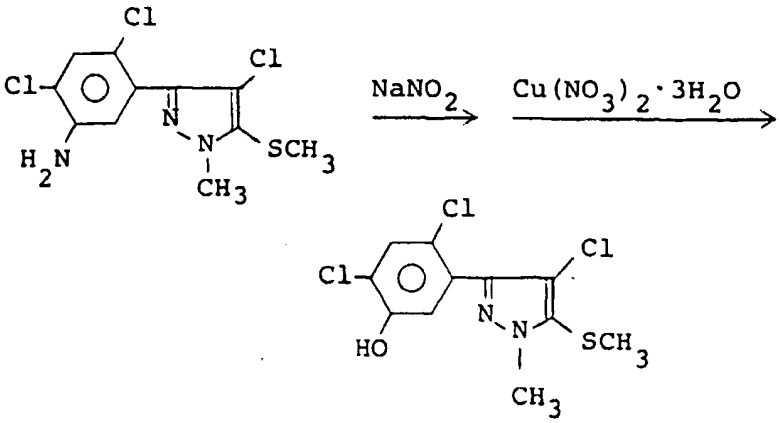
Example 7 Preparation of 3-(5-amino-4-chloro-2-fluorophenyl)-4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazole (compound No. 405)



To 4-chloro-3-(4-chloro-2-fluoro-5-nitrophenyl)-5-difluoromethoxy-l-methyl-1H-pyrazole (3.0 g, 8.5 mmoles) in a mixture of 15 ml ethanol and 15 ml conc. hydrochloric acid was added stannous chloride (7.70 g, 34 mmole) and the reduction was conducted under reflux for 8 hours. Then the product solution was poured into ice-cold water, the resulting mixture was made alkaline with 20% aqueous NaOH, and the objective product was extracted with ethyl acetate. Dehydration and concentration of the extract solution and purification of the residue by column chromatography gave the title compound (2.15 g), yield 78%.

NMR, δ (ppm): 3.73 (3H, s), 3.85 (2H, s, br), 6.64 (1H, t, J=72 Hz), 6.82 (1H, d, J=8 Hz), 7.04 (1H, d, J=10 Hz).

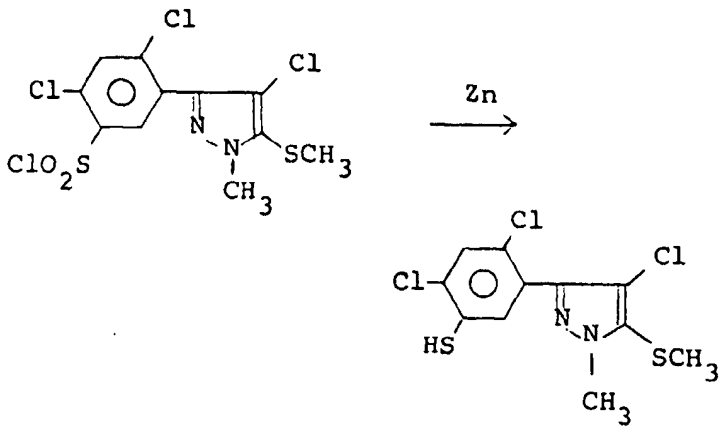
Example 8 Preparation of 4-chloro-3-(2,4-dichloro-5-hydrophenyl)-1-methyl-5-methylthio-1H-pyrazole (compound No. 2)



A solution of sodium nitrile (1.33 g) in 4.5 ml water was added dropwised to a 5°C or lower temperature solution of 4-chloro-3-(5-amino-2,4-dichlorophenyl)-1-methyl-5-methylthio-1H-pyrazole (5.67 g, 17 mmoles) in 20 ml 50% sulfuric acid while maintaining the reaction temperature between -5 and +5°C. After the addition was finished, the reaction mixture was further stirred for 20 minutes at 0 - 5°C. Then the resulting solution was removed into a 1-ℓ Elenmeyer flask, and a 40% aqueous copper nitrate solution (about 100 g of a total of 318.8 g) was first added dropwise to the former solution kept at temperatures of 0 - 10°C, and secondly the remainder copper nitrate solution was added dropwise. After stirring of the whole mixture for 30 minutes at room temperature, cuprous oxide (2.51 g, 17 mmoles) was added in limited amounts, conducting the reaction for 1 hour at room temperature. Then the resulting solution was poured into ice-cold water, and the objective product was extracted with ethyl acetate. The extract solution was washed with water and concentrated. The residue was purified by recrystallization from methylene chloride, giving crystals

(2.61 g) of the title compound, m.p. 163.2°C, yield 46.4%.

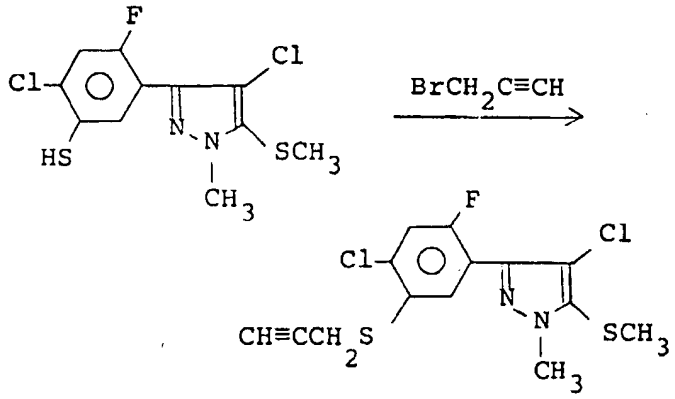
Example 9 Preparation of 4-chloro-3-(2,4-dichloro-5-mercaptophenyl)-1-methyl-5-methylthio-1H-pyrazole (compound No. 15)



A mixture of 4-chloro-3-(5-chlorosulfonyl-2,4-dichlorophenyl)-1-methyl-5-methylthio-1H-pyrazole (7.71 g, 19 mmoles), 80 ml glacial acetic acid, and zinc dust (24.8 g, 380 mmoles) was subjected to reaction under reflux for 3.5 hours. The resulting solution was poured into ice-cold water, and the objective product was extracted with ethyl acetate. The extract solution was washed with water, dehydrated, and concentrated, giving the title compound (5.75 g) as oily matter, yield 89.1 %.

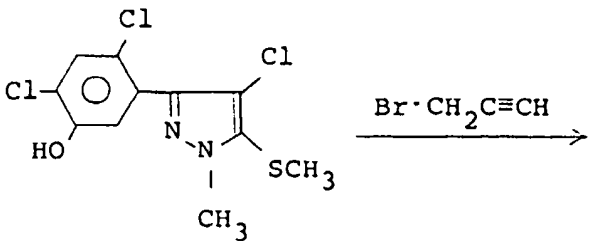
nD (24.3°C): 1.6303

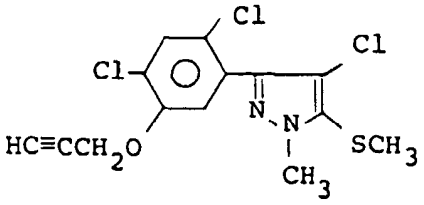
Example 10 Preparation of 4-chloro-3-[4-chloro-2-fluoro-5-(2-propynylthio)phenyl]-1-methyl-5-methylthio-1H-pyrazole (compound No. 221)



A mixture of 4-chloro-3-(4-chloro-2-fluoro-5-mercaptophenyl)-1-methyl-5-methylthio-1H-pyrazole (0.60 g, 1.86 mmoles), 30 ml acetone, a K_2CO_3 powder (2.04 mmoles), and propargyl bromide (2.23 mmoles), was subjected to reaction under reflux for 2 hours. Then the resulting solution was filtered to remove acetone-insoluble matter and the filtrate was concentrated and purfied by silica gel column chromatography, giving crystals (0.54 g) of the title compound, m.p. 93 - 96°C, yield 80.5%.

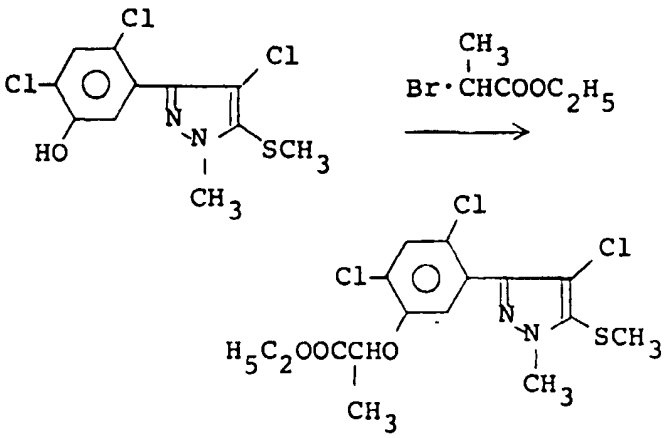
Example 11 Preparation of 4-chloro-3-[2,4-dichloro-5-(2-propynyloxy)phenyl]-1-methyl-5-methylthio-1H-pyrazole (compound No. 5)





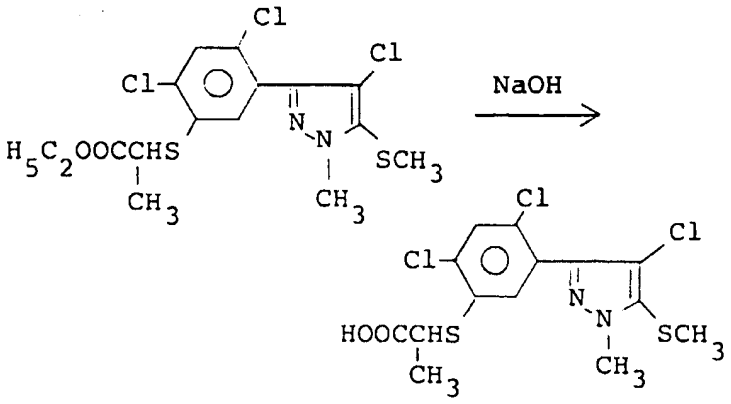
A mixture of 4-chloro-3-(2,4-dichloro-5-hydroxyphenyl)-1-methyl-5-methylthio-lH-pyrazole (2.00 g, 6.2 mmoles), 20 ml acetone, anhydrous K_2CO_3 (1.28 g, 9.3 mmoles), and propargyl bromide (1.10 g, 9.3 mmoles) was subjected to reaction under reflux for 2 hours. Then the resulting solution was filtered to remove acetone-insoluble matter. The filtrate was concentrated and purified by silica gel column chromatography, giving crystals (1.89 g) of the title compound, m.p. 71.5 - 72.5°C, yield 84.3%.

Example 12 Preparation of ethyl 2-[5-(4-chloro-1-methyl-5-methylthio-lH-pyrazol-3-yl)-2,4-dichloro-phenoxy] propionate (compound No. 11)



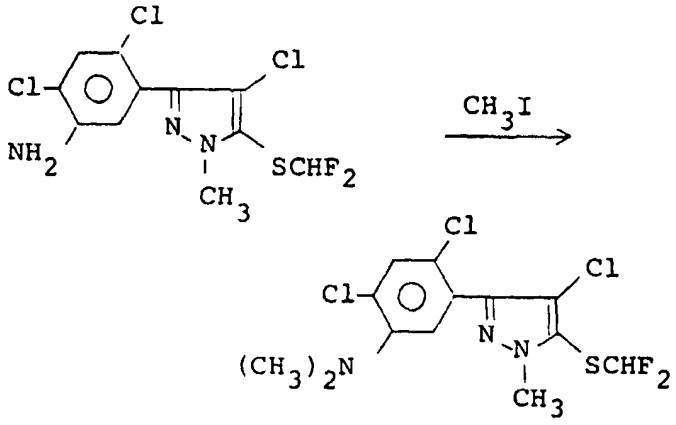
A mixture of 4-chloro-3-(2,4-dichloro-5-hydroxyphenyl)-l-methyl-5-methylthio-1H-pyrazole (3.50 g, 10.8 mmoles), 50 ml acetone, anhydrous K_2CO_3, and ethyl 2-bromopropionate (2.06 g, 11.4 mmoles) was subjected to reaction under reflux for 2 hours. Then the resulting solution was filtered to remove acetone-insoluble matter, and the filtrate was concentrated and purified by silica gel column chromatography, giving the title compounds as oily matter, yield 70.0%. nD(28.8°C):1.5763

Example 13 Preparation of 2-[5-(4-chloro-l-methyl-5-methylthio-1H-pyrazol-3-yl)-2,4-dichlorophenylthio]propionic acid (compound No. 25)



After addition of NaOH (0.31 g, in pellet form, 95% content) to a solution of ethyl 2-[5-(4-chloro-l-methyl-5-methylthio-1H-pyrazol-3-yl)-2,4-dichlorophenylthio]propionate (2.28 g, 5.18 mmoles) in 50 ml ethanol, several drops of water added, thereby conducting the reaction for 3 hours at room temperature. Then the solvent was removed by evaporation of the resulting solution under reduced pressure. Water and ethyl acetate were added to the residue and the formed aqueous layer was separated. The aqueous layer was acidified with hydrochloric acid. The objective product was extracted from the aqueous layer with ethyl acetate. The extract solution was washed with water, dehydrated, and evaporated to dryness, giving crystals (1.74 g) of the title compound, m.p. 180.0°C, yield 81.6%.

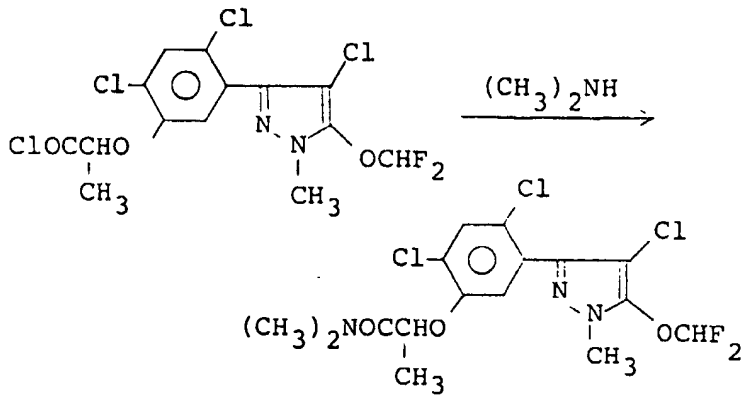
Example 14 Preparation of 4-chloro-3-(2,4-dichloro-5-dimethylaminophenyl)-5-difluoromethylthio-1-methyl-1H-pyrazole (compound No. 207)



A mixture of 3-(5-amino-2,4-dichlorophenyl)-4-chloro-5-difluoromethylthio-1-methyl-1H-pyrazole (0.72 g, 2 mmoles), 15 ml sulforane, $NaHCO_3$ (0.18 g, 2.2 mmoles), and methyl iodide (0.34 g, 2.4 mmoles) was subjected to reaction at 80°C for 16 hours. Then the resulting mixture was poured into water and the objective product was extracted with ether. The extract solution was washed with water, dehydrated, and evaporated to dryness, giving the title compound (0.62 g) in paste form, yield 80.2%.

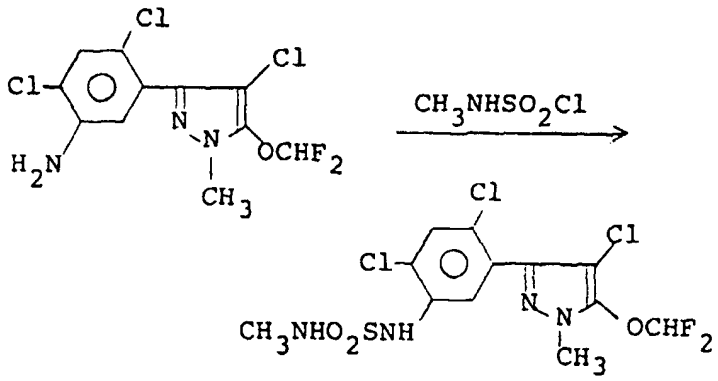
nD (21.6°C): 1.5838

Example 15 Preparation of N,N-dimethyl 2-[5-(4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazol-3-yl)-2,4-dichlorophenoxy]propionamide (compound No. 110)



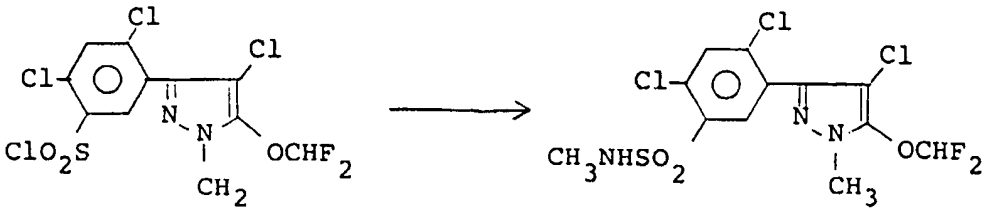
A solution of 2-[5-(4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazole-3-yl)-2,4-dichlorophenoxy]propionyl chloride (0.80 g, 1.78 mmoles) in 20 ml anhydrous tetrahydrofuran was added dropwise to a solution of 50% dimethylamine (0.32 g, 3.56 mmoles) in 20 ml tetrahydrofuran at room temperature to react the pyrazole derivative with dimethylamine. After the addition was finished, the reaction was continued further for 1 hour. Then ethyl acetate was added to the reaction product solution, and the mixture was washed with water. The separated organic layer was dehydrated, concentrated under reduced pressure, and purified by silica gel chromatography, giving crystals (0.24 g) of the title compound, m.p. 148.9°C, yield 27%.

Example 16 Preparation of 4-chloro-3-(2,4-dichloro-5-methylaminosulfonylaminophenyl)-5-difluoromethoxy-1-methyl-1H-pyrazole (compound No. 149)



N-Methylaminosulfonyl chloride (0.22 g, 1.67 mmoles) was added dropwise to a solution of 3-(5-amino-2,4-dichlorophenyl)-4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazole (0.44 g, 1.28 mmoles) and triethylamine (0.17 g, 1.67 mmoles) in 20 ml anhydrous tetrahydrofuran under cooling with ice to react with the pyrazole derivative. After the addition was finished, the reaction was continued further for 2 hours. Then ethyl acetate was added to the reaction product solution, and the mixture was washed with water. The separated organic layer was dehydrated, concentrated, and purified by recrystallization from a n-hexane-ether mixture, giving the title compound (0.40 g), m.p. 133.0°C, yield 71.7%.

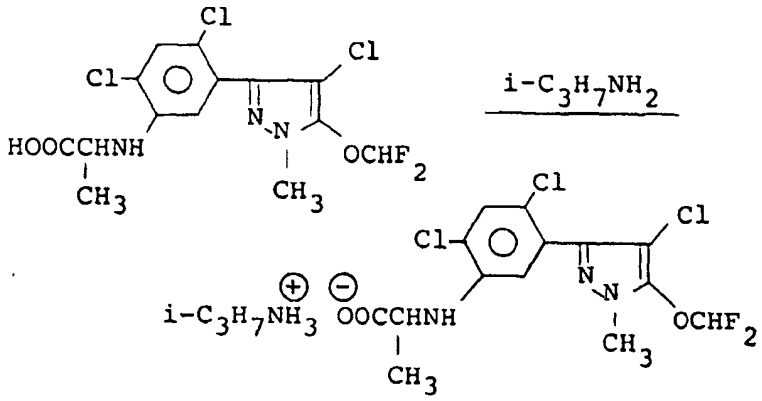
Example 17 Preparation of 5-(4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazol-3-yl)-N-methyl-2,4-dichlorobenzenesulfonamide (compound No. 164)



A solution of 4-chloro-3-(5-chlorosulfonyl-2,4-dichlorophenyl)-5-difluoromethoxy-1-methyl-1H-pyrazole (0.43 g, 1 mmole) in 2 ml tetrahydrofuran was added dropwise to a 40% aqueous methylamine solution dissolved in 20 ml tetrahydrofuran to react the pyrazole derivative with methylamine. After the addition was finished, the reaction was continued further for 30 minutes at room temperature. Then ethyl acetate was added to the reaction product solution to extract the objective product. The extract solution was washed with water, dehydrated, and evaporated to dryness, giving the title compound (0.42 g) in paste form, yield 100%.

nD (17.9°C): 1.5461

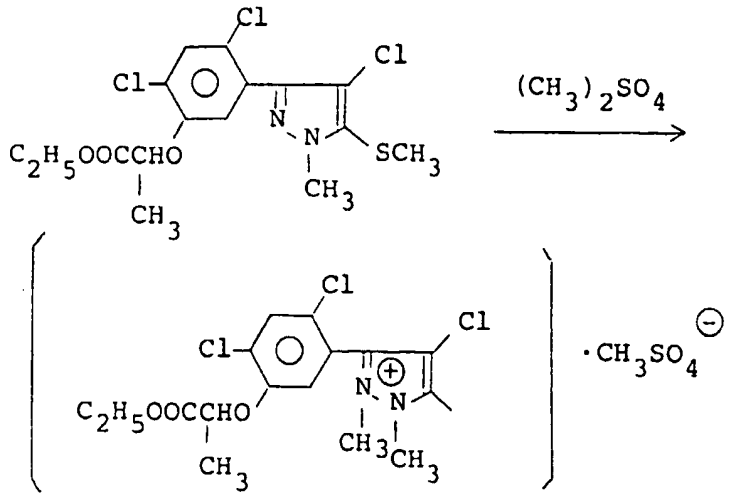
Example 18 Preparation of 2-[5-(4-chloro-5-difluoromethoxy-1-methyl-1H-pyrazol-3-yl)-2,4-dichloroanilino] isopropylammonium propionate (compound No. 160)



Isopropylamine (0.05 g, 0.88 mmole) was added to a solution of 2-[5-(4-chloro-5-difluoromethoxy-l-methyl-lH-pyrazol-3-yl)-2,4-dichloroanlino]-propionic acid (0.33 g, 0.8 mmole) in 20 ml tetrahydrofuran, thereby being reacted with the acid for 30 minutes at room temperature. Thereafter the solvent was removed from the reaction product solution by evaporation under reduced pressure, giving the title compound quantitatively (100% yield) as oily matter.

NMR δ (ppm); 1.12 (6H, d), 1.48 (3H, d), 2.60 - 3.30 (1H, m), 3.73 (3H, s), 3.91 (1H, q), 6.55 (1H, s), 6.68 (1H, s) 7.73 (1H, s), 7.98 (3H, s).

Example 19 Preparation of ethyl 2-[5-(4-chloro-1,2-dimethyl-5-methylthio-1H-pyrazolium-3-yl)-2,4-dichlorophenoxy] propionatemethyl sulfate (compound No. 215)



Dimethyl sulfate (0.13 g, 1.04 mmoles) was added to a solution of ethyl 2-[5-(4-chloro-l-methyl-5-methylthio-1H-pyrazol-3-yl)-2,4-dichlorophenoxy]-propionate (0.22 g, 0.52 mmole) in 20 ml benzene, thereby being reacted with the pyrazole derivative under reflux for 5 days. Thereafter the solvent was removed from the reaction product solution by evaporation under reduced pressure. The residue was dissolved in ethyl acetate, the solution was washed with a small amount of water, and the organic layer was dehydrated and evaporated to dryness, giving the title quateranry salt in paste form, yield 52.6%.

nD (25.7°C): 1.5548

The present inventive 3-(substituted phenyl)-pyrazole derivatives represented by formula (I) and salts thereof are capable of controlling annual and perennial weeds grown in paddy fields, upland fields, orchards, and swamps, such as barnyardgrass (Echinochloa Crus-galli Beauv., an annual gramineous grass which is a typical weed strongly injurious, grown in paddy fields), Mizugayatsuri (Cyperus serotinus Rottb., a perennial weed of Cyperaceae family, grown in swamps, ditches and paddy fields), Urikawa (Sagittaria pygmaea Miq., an injurious perennial weed of Alismataceae family, grown in swamps, ditches, and paddy fields), Hotarui (Scirpus juncoides Roxb. subsp. juncoides, a perennial cyperaceous weed grown in swamps, water ways, and paddy fields), wild oats (Avena fatua L., an annual gramineous grass grown in plains, highlands, and upland fields), large crabgrass (Digitaraia adscendcus Henr., an annual gramineous grass which is a typical strongly injurious weed grown in upland fields and orchards), Curlydock (Rumex japonicus Houtt, a perennial polygonaceous weed grown in upland fields and on roadsides), umbrella sedge (Cyperus Iria L., an annual cyperaceous weed grown in upland fields and on roadsides), Redroot pigweed (Amaranthus vetroflexus L., an annual weed of Amaranthaceae family grown in upland fields, roadsides, and vacant lands), Cleavers (Galium aparine L, a strongly injurious annual weed of Rubiaceae family grown in upland fields), Bridseye Speedwell (Veronica persica L., a strongly injurious weed of Scrophulariaceae family grown in upland fields and orchards), Scented mayweed (Matricaria chamomilla L., injurious composite weed grown in upland fields), velvetleaf (Abutilon theophrasti L., a strongly injurious weed of Malvaceae family grown in upland fields), cocklebur (Xanthium strumarium L., a strongly injurious annual composite weed grown in upland fields), and tall morningglory (Ipomoea purpurea Voigt, a strongly injurious weed of Convolvulaceae family grown in upland fields).

Since the 3-(substituted phenyl)pyrazole derivatives of formula (I) and salts thereof exhibit excellent controlling effect on weeds before or immediately after germination, characteristic physiological activities of these compounds can be manifested by treating fields with the derivative or the salt before or after planting of useful plants therein (including fields where useful plants are already planted) in the stage prior to weed emergence or in the period from the initial stage to the middle stage of weed growth. However, the applications of the present inventive herbicides are not limited such forms as stated above. The present herbicides can be applied to control not only weeds in paddy fields or upland fields but also general weeds grown in other places, for example, reaped fields, temporarily non-cultivated paddy fields and upland fields, ridges between paddy fields, agricultural pathways, waterways, lands constructed for pasture, graveyards, parks, roads, playgrounds, unoccupied areas around buildings, reclaimed lands, railways, and forests. It is most desirable in economy as well to treat these areas with the present herbicides, before the end of the

initial stage of weed growth, but the treatment is not limited to this but can be carried out in the middle stage of weed growth.

The 3-(substituted phenyl)pyrazole derivative of formula (I) or salt thereof, when applied as a herbicide, is generally made up, according to the customary procedure for preparing agricultural chemicals, into a form convenient to use. That is, the pyrazole derivative or salt thereof is blended with a suitable inert carrier and if necessary, further with an adjuvant, in proper ratios, and the mixture is made up into a suitable form of preparation, e.g. a suspension, emulsifiable concentrate, solution, wettable powder, granules, dust, or tablets, through dissolution, disersion, suspension, mixing, impregnation, adsorption, or sticking. In the present invention, either solid or liquid inert carriers may be used. Suitable materials as solid carriers include soybean flour, cereal flour, wood flour, bark flour, saw dust, powdered tobacco stalks, powdered walnut shells, bran, powdered cellulose, extraction residues of vegetables, powdered synthetic polymers or resins; clays (e.g. kaolin, bentonite, and acid clay), talcs (e.g. talc and pyrophyllite), silica powders or flakes (e.g. diatomaceous earth, silica sand, mica and white carbon, i.e. highly dispersed silicic acid, also called finely divided hydrated silica or hydrated silicic acid), activated carbon, powdered sulfur, powdered pumice, calcined diatomaceous earth, ground brick, fly ash, sand, calcium carbonate powder, calcium phosphate powder, and other inorganic or mineral powders, chemical fertilizers (e.g. ammonium sulfate, ammonium nitrate, urea, and ammonium chloride) and compost. These materials may be used alone or in combination.

Suitable materials as liquid carriers include liquids which themselves have solvent power as well as liquids which do not have solvent power but can disperse active ingredients with the aid of adjuvants. Typical examples of such liquids, which may be used alone or in combination, are water, alcohols (including methanol, ethanol, isopropanol, butanol, and ethylene glycol), ketones (including acetone, methyl ethyl ketone, methyl isobutyl ketone, di-isobutyl ketone, and cyclohexanone), ethers (including ethyl ether, dioxane, Cellosolve, dipropyl ether, and tetrahydrofuran), aliphatic hydrocarbons (including gasoline, and mineral oils), aromatic hydrocarbons (including benzene, toluene, xylene, solvent naphtha, and alkylnaphthalenes), halogenated hydrocarbons (including dichloroethane, chloroform and carbon tetrachloride), esters (including ethyl acetate, diisopropyl phthalate, dibutyl phthalate, and dioctyl phthalate), amides (including dimethylformamide, diethylformamide, and dimethylacetamide), nitriles (including acetonitrile), and dimethylsulfoxide.

The following materials are cited as typical examples of the adjuvants which may be used alone or in combination or may not be used at all.

For the purpose of emulsifying, dispersing, solubilizing, and/or wetting active ingredients, there may be used surface active agents such as polyoxyethylene alkyl ethers, polyoxyethylene alkylaryl ethers, polyoxyethylene higher fatty acid esters, polyoxyethylene resinates, polyoxyethylene sorbitan monolaurate, polyoxyethylene sorbitan monooleate, alkylarylsulfonates, naphthalenesulfonic acid condensation products, ligninsulfonates, and higher alcohol sulfate esters.

For the purpose of stabilizing the dispersion of active ingredients, tackifying and/or binding them, there may be used adjuvants, for example, casein, gelatin, starch, methylcellulose, carboxymethylcellulose, gum arabic, polyvinyl alcohol, turpentine, bran oil, bentonite, and ligninsulfonates.

For the purpose of improving flow properties of solid herbicidal products there may be used adjuvants, for example, waxes, stearates, and alkyl phosphates.

Adjuvants, e.g. naphthalenesulfonic acid condensation products and polyphates, can be used as peptizers in dispersible herbicidal products.

Adjuvants, e.g. silicone oils, can also the used as defoaming agents.

The content of active ingredients may be varied as occasion demands; the suitable contents are from 0.01 to 50% by weight for preparing, for example, dusts or granules as well as emulsifiable concentrates or wettable powders.

For destroying various weeds or inhibiting their growth, the herbicidal composition containing the 3-(substituted phenyl)pyrazole derivative of formula (I) or salt thereof as an active ingredient is applied as such or after being properly diluted with or suspended in water or other media, in amounts effective for destroying weeds or inhibiting their growth, to the foliage and stalks of the weeds or to soil in the area where the emergence or growth of weeds is undesirable.

The amount of the present herbicidal composition to be used, depending upon various factors, e.g. the purpose of application, object weeds, emergence or growth states of weeds and crops, emergence tendency of weeds, weather, environmental conditions, form of the herbicidal composition, mode of the application, type or state of the application locus, and time of the application, is chosen properly according to the purpose from the range of 1.0 g to 10 Kg, in terms of the amount of active ingredient, per hectare.

When the present herbicidal composition is applied to paddy fields or upland fields, it is desirable to choose such low doses as not to injure crops but to destroy weeds or control their growth. When the composition is applied to non-farming areas, suitable doses of active ingredient for destroying the weeds are chosen from amounts of 100 g/hectare and more.

The present herbicidal composition can be applied jointly with other herbicides for the purpose of expanding both the range of controllable weed species and the period of time when effective applications are possible or for the purpose

of reducing the dosage.

The following examples illustrate herbicidal effects and formulations of the present inventive herbicidal composition without limiting the scope of the invention.

Test Example 1 Herbicidal effect on paddy field weeds of pre-emergence stage

Pots (1/10,000-are) were filled with soil to simulate a paddy field, then plated separately with seeds of barnyardgrass, konagi (Monochoria vaginalis (Burm.f.) Presl var plantaginae), and hotarui which are all injurious weeds grown in paddy fields, and were conditioned so that the seeds might become in pre-emergence stage.

Soil in each pot was sprayed with each of solutions containing compounds (listed in Table 1) of the present invention as active ingredients at a predetermined concentration. 21 Days later, the herbicidal effect was examined, the percentages of killed weeds were calculated in comparison with those on the untreated soil, and the herbicidal activity was judged and the chemical injury of a rice plant was examined at the same time and judged according to the following criterion:

° Herbicidal activity

Rating	Percentage of killed weed
5	95% or more
4	70 - 95% (exclusive)
3	50 - 70% (exclusive)
2	30 - 50% (exclusive)
1	10 - 30% (exclusive)
0	less than 10%

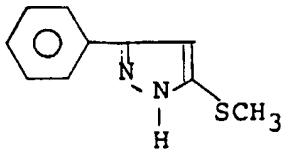
° Phytotoxicity

Rating	Degree of phytotoxicity
0	No phytotoxicity
1	Browning occurs but disappears in the initial growth stage, growth inhibition is not observed.
2	Browning and distinct growth inhibition are observed but normal conditions are soon restored.
3	Browning and growth inhibition are remarkable and the restoration is slow.
4	Browing and growth inhibition are remarkable and some of the rice plants are killed.
5	All the rice plants were killed.

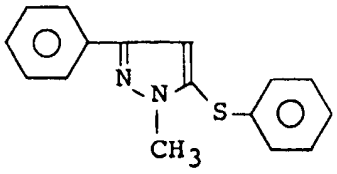
For comparison the following compounds were also tested.

Compound A: 3-phenyl-5-methylthiopyrazole, described on page 3 in Japanese Patent Application Kokai No. Sho. 51-91861; compound B: described in Example 1 on page 4 in the same patent application; compound C: compound No. 8 described in Japanese Patent Application Kokai No. Sho. 54-70270; compound D: compound No. 159 described on page 9 in Japanese Patent Application Kokai No. Sho. 55-9062.

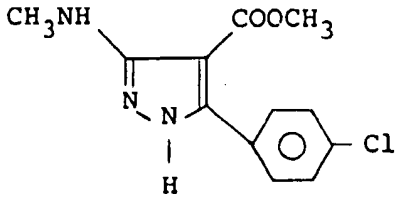
Compound A.



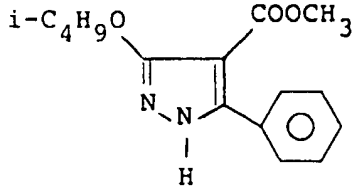
Compound B.



Compound C.



Compound D.



Results of the test are shown in Table 5.

Table 5

		Phytotoxicity	Pre-emergence treatment		
Comp'd No.	Amount of active ingredient Kg/ha	rice	Barnyardgrass	Hatarui	Konagi
86	0.8	5	5	5	5
	0.1	4	5	5	5
87	0.8	5	5	5	5
	0.1	4	5	5	5
117	0.8	4	5	5	5
	0.1	3	5	5	5
119	0.8	4	5	5	5
	0.1	2	5	5	5
152	0.8	3	5	5	5
	0.1	2	5	5	5
153	0.8	5	5	5	5
	0.1	3	5	5	5
253	0.8	5	5	5	5
	0.1	1	3	5	5
256	0.8	5	5	5	5
	0.1	4	5	5	5
257	0.8	5	5	5	5
	0.1	3	5	5	5
258	0.8	5	5	5	5
	0.1	4	5	5	5
259	0.8	5	4	5	5
	0.1	3	2	4	4
260	0.8	5	5	5	5
	0.1	3	2	5	5
268	0.8	5	5	5	5
	0.1	3	5	5	5
269	0.8	5	5	5	5
	0.1	3	2	5	5
275	0.8	5	5	5	5
	0.1	2	3	5	5
344	0.8	5	5	5	5
	0.1	4	5	5	5
346	0.8	5	5	5	5
	0.1	4	5	5	5
359	0.8	5	5	5	5
	0.1	3	5	5	5

Table 5 (continued)

		Phytotoxicity	Pre-emergence treatment		
Comp'd No.	Amount of active ingredient Kg/ha	rice	Barnyardgrass	Hatarui	Konagi
368	0.8	5	5	5	5
	0.1	2	5	5	5
369	0.8	5	5	5	5
	0.1	3	5	5	5
370	0.8	5	5	5	5
	0.1	2	2	5	5
371	0.8	5	5	5	5
	0.1	2	1	5	5
A	5	0	0	0	0
B	5	0	0	2	0
C	5	0	0	0	0
D	5	0	0	0	0

As shown in table 5, the present 3-(substituted phenyl) pyrazole derivatives or salt thereof, represented by the general formula (I), exhibit more excellent controlling effects of weeds than comparative compound A, B, C or D at pre-emergence treatment in paddy fields. Even when the derivatives exhibit both herbicidal activity on weeds and phytotoxicity on crops at the same dosage, however, by the selection of a proper lower dosage, sufficient herbicidal activity is retained but phytotoxicity on crops is reduced remarkably.

Test Example 2 Herbicidal effect on paddy field weeds of post-emergence stage

Pots (1/10,000-are) were filled with soil to simulate a paddy field, then planted separately with seeds of barnyardgrass and hotarui and with tubers of mizugayatsuri and urikawa, which are all injurious weeds grown in paddy fields, and were conditioned so that these weeds might grow to 1-leaf stage.

Soil in each pot was sprayed with each of solutions containing compounds (listed in Table 1) of the present invention as active ingredient at a predetermined concentration. 21 Days later, the herbicidal effect was examined, the percentages of killed weeds were calculated in the same manner as in Test Example 1, and the phytotoxicity of a rice plant also examined and judged according the criterior shown in Test Example 1.

Results of the test are shown in Table 6.

Table 6

Comp'd No.	Amount of active ingredient Kg/ha	Phyto-toxicity rice	Effect of past-emergence treatment			
			Barnyardgrass	Hotarui	Mizugaytsuri	Urikawa
2	5	1	4	1	0	0
3	5	2	5	4	2	3
4	5	5	5	5	3	4
	0.8	3	5	3	1	2
5	5	5	5	5	5	5
	0.8	4	5	4	4	3
6	5	3	5	4	5	3
7	5	4	5	4	3	1
	0.8	2	4	1	0	1
8	5	3	5	4	1	2
9	5	5	5	5	5	5
	0.8	4	5	4	5	5
10	5	5	5	5	5	5
	0.8	5	5	4	5	5
	0.1	2	5	2	3	4

- to be cont'd -

11	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	4	4	5	5	4	5	5	4	5	5	5	4	5	5	5	5	5	5	5	5	5	5
	0.1	2	3	5	5	2	5	5	1	5	5	5	4	5	5	5	5	5	5	5	5	5	5
12	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	4	4	5	5	4	5	5	4	5	5	5	4	5	5	5	5	5	5	5	5	5	5
	0.1	2	3	5	5	2	5	5	1	5	5	5	4	5	5	5	5	5	5	5	5	5	5
13	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	4	4	5	5	4	5	5	4	5	5	5	4	5	5	5	5	5	5	5	5	5	5
	0.1	2	3	5	5	2	5	5	1	5	5	5	4	5	5	5	5	5	5	5	5	5	5
14	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	4	4	5	5	4	5	5	4	5	5	5	4	5	5	5	5	5	5	5	5	5	5
16	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
18	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
19	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
20	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
21	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
23	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
25	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	4	4	5	5	4	5	5	4	5	5	5	4	5	5	5	5	5	5	5	5	5	5
26	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	4	4	5	5	4	5	5	4	5	5	5	4	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

27	5	4	5	5	4	5
	0.8	3	3	2	3	3
28	5	5	5	5	2	5
	0.8	2	5	2	0	4
29	5	5	5	5	4	5
	0.8	3	5	3	2	4
30	5	5	5	5	2	5
	0.8	2	5	2	0	4
31	5	5	5	5	4	5
	0.8	3	5	2	1	4
32	5	5	5	5	2	5
	0.8	2	5	2	0	4
33	5	5	5	5	2	5
	0.8	2	5	2	0	4
34	5	5	5	5	2	5
	0.8	2	5	2	0	4
35	5	5	5	5	2	5
	0.8	2	5	2	0	4
37	5	5	5	4	5	5
	0.8	2	5	2	2	5

- to be cont'd -

38	5	5	2	4	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
41	5	4	1	2	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
42	5	2	1	2	0	4	4	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5
43	5	5	1	2	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
44	5	4	1	2	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
45	5	4	1	2	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
46	5	4	1	2	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
47	5	4	1	2	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
48	5	4	1	2	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
49	5	5	1	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
50	0.8	5	3	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
51	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
52	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
53	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
54	5	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

55	5	2	2	2	5	2	5	2	2	5	2	2	5	2	2	5	2	2	5	2	2	4	1	5	2
57	5	0	2	0	2	2	0	2	0	5	2	0	5	2	0	5	2	0	5	2	0	5	3	5	5
59	5	2	5	4	2	3	3	5	3	5	2	3	5	2	3	5	2	3	5	2	3	5	3	5	5
62	5	5	5	2	0	3	3	5	3	5	2	3	5	2	3	5	2	3	5	2	3	5	3	5	5
63	0.8	1	2	0	1	2	0	1	0	2	1	0	2	1	0	2	1	0	2	1	0	2	1	2	3
64	5	5	5	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
66	0.8	2	2	3	1	3	2	5	2	3	2	3	5	2	3	5	2	3	5	2	3	5	2	3	5
67	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
68	0.8	1	2	3	1	3	1	5	2	3	3	1	5	2	3	5	2	3	5	2	3	5	2	3	5
69	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
71	0.8	2	2	3	1	3	2	5	2	3	3	1	5	2	3	5	2	3	5	2	3	5	2	3	5
72	0.8	3	5	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

73	5	5	4	4	4	4	2	2	4	4	4	4	2	2	4	4	4	4	2	2	4	4
74	5	5	4	4	5	5	4	5	5	4	4	4	4	5	5	4	4	4	4	5	5	4
	0.8	0.8	1	0	3	3	1	3	3	0	0	1	0	3	3	0	0	1	0	3	3	0
75	5	5	2	4	0	2	4	2	4	4	4	2	4	2	4	4	4	2	4	2	4	4
76	5	5	4	5	4	2	4	2	4	5	5	4	5	2	4	5	5	4	5	2	4	5
77	5	5	1	1	5	2	4	2	4	5	5	4	5	2	4	5	5	4	5	2	4	5
78	5	5	2	2	1	1	4	2	4	5	5	4	5	2	4	5	5	4	5	2	4	5
81	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
82	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
83	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
84	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
85	5	5	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
86	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0.1	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
87	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0.1	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
88	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3

- to be cont'd -

89	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
90	0.8	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
91	0.8	5	5	3	5	5	5	5	5	0	5	5	5	5	5	5	5	5	5	5	5
92	0.1	0.8	5	3	5	4	5	5	5	2	5	4	5	5	5	5	5	4	5	5	3
93	0.8	0.1	5	3	5	3	5	4	5	2	5	5	5	5	5	5	5	4	5	5	4
94	0.1	0.8	5	3	5	3	5	3	5	2	5	3	5	4	5	4	5	2	5	5	2
95	0.1	0.8	5	3	5	3	5	3	5	2	5	3	5	4	5	4	5	2	5	5	2
96	5	0.8	5	3	5	3	5	3	5	2	5	3	5	4	5	4	5	2	5	5	2
97	0.8	0.8	5	3	5	3	5	3	5	2	5	3	5	4	5	4	5	2	5	5	2
98	0.1	0.8	5	3	5	3	5	3	5	2	5	3	5	4	5	4	5	2	5	5	2
	0.1	0.8	5	3	5	3	5	3	5	2	5	3	5	4	5	4	5	2	5	5	2

- to be cont'd -

99	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
100	0.8	5	5	5	5	0	5	5	5	1	5	0	5	1	5	0	5	5	5	5	
101	0.8	5	5	5	5	1	5	5	5	0	5	0	5	4	5	0	5	4	5	5	
102	0.1	5	5	5	5	3	5	5	5	0	5	0	5	4	5	4	5	4	5	5	
103	5	3	5	4	5	2	5	4	5	2	5	2	5	4	5	3	5	3	5	3	
104	0.8	5	0.8	0.8	0.1	5	0.8	0.8	0.1	0.8	0.1	0.8	0.1	0.8	0.1	5	0.1	5	0.1		
105	0.8	5	0.8	0.8	0.1	5	0.8	0.8	0.1	0.8	0.1	0.8	0.1	0.8	0.1	5	0.1	5	0.1		
106	0.1	5	0.8	0.8	0.1	5	0.8	0.8	0.1	0.8	0.1	0.8	0.1	0.8	0.1	5	0.1	5	0.1		
108	5	5	0.8	0.8	0.1	5	0.8	0.8	0.1	0.8	0.1	0.8	0.1	0.8	0.1	5	0.1	5	0.1		
109	5	5	0.8	0.8	0.1	5	0.8	0.8	0.1	0.8	0.1	0.8	0.1	0.8	0.1	5	0.1	5	0.1		

- to be cont'd -

110	5	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
111	5	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
112	5	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
113	5	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
114	5	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
115	5	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
116	5	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
117	5	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
118	5	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
119	5	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

120	5	5	5	5	5	3	5	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5
121	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
122	0.8	3	3	4	3	5	3	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5
123	5	5	5	5	5	5	3	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5
125	0.8	5	5	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
126	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
127	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
128	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
129	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

129	0.1	5	4	5	5	5	5	5	5
130	0.8	5	5	5	5	5	5	5	5
	0.1	2	5	5	2	5	5	5	5
131	5	5	5	5	5	5	5	5	5
	0.8	3	2	5	0	5	5	5	5
132	5	5	5	5	5	5	5	5	5
	0.8	3	3	4	1	5	5	5	5
133	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5
	0.1	3	0	5	5	5	5	5	5
134	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5
	0.1	3	5	5	0	5	5	5	5
135	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5
	0.1	3	4	5	3	5	5	5	5
136	5	5	4	5	5	5	5	5	5
	0.8	5	3	5	5	5	5	5	5
	0.1	3	2	5	2	5	5	5	5
137	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5

- to be cont'd -

137	0.1	3	5	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
138	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
139	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
140	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	3	5	5	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
141	5	5	5	5	5	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
142	0.1	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	5	5	5	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
143	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
144	5	4	5	5	5	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

145	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
146		5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
147		5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
148		5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
149	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
		5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
150	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
		5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
151	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
		5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
152	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
		5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
153	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
		5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
154	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

155	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8		0.8		0.8		0.8		0.8		0.8		0.8	0.1		0.8	0.1		0.8		0.1	
156	5	5	5	5	5	5	3	1	2	0	2	0	5	4	2	5	5	5	5	3		
157	5	2	5	2	5	5	5	4	5	3	3	0	5	5	4	5	5	3	5	5		
158	5	4	5	4	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5		
159	5	3	5	2	5	3	5	3	5	3	5	3	5	5	2	5	4	3	5	3		
160	5	0.8	5	0.8	5	0.8	5	0.8	5	0.8	5	0.8	5	0.8	0.1	5	0.8	0.1	0.8	0.1		
161	5		5		5		5		5		5		5			5						
162	5		5		5		5		5		5		5			5						
163	5		5		5		5		5		5		5			5						

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164	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	3	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
165	5	4	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
166	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
167	5	2	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
168	5	4	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
	0.8	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
169	5	2	4	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
	0.8	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
170	5	2	0	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
	0.8	0	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
171	5	4	5	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
172	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
173	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
174	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
175	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
176	5	1	5	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
177	5	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3

- to be cont'd -

178	5	5	2	3	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
179	5	5	5	4	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	2	2	0	2	2	2	0	2	2	2	2	2	2	2	2	2	2	2	2	2	2
180	5	5	5	2	2	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
181	5	5	5	5	4	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
182	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	5	5	5	4	5	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
183	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
184	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
185	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
186	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
187	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
188	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
189	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
190	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
191	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

192	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
193	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
194	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
195	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
196	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
197	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
198	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
199	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
200	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
201	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5
202	5	0.8	5	5	3	3	5	5	4	5	5	5	4	5	3	5	5	5	5	5	5	5	5

- to be cont'd -

203	5	0.8	5	5	5	5	5	5
204	5	0.8	5	4	2	2	2	2
205	5		5	5	4	4	5	5
206	5		5	5	5	5	5	5
207	5	0.8	5	5	5	3	1	1
208	5		5	5	5	2	2	5
209	5		5	5	5	4	2	5
210	5	0.8	5	5	5	4	5	5
211	5		5	5	5	1	3	5
212	5	0.8	5	5	5	5	2	5
213	5		5	5	5	4	5	5
214	5		5	5	5	4	5	5
216	5		5	5	5	4	5	5
218	5	0.8	5	5	5	5	5	5

- to be cont'd -

220	5	0.8	5	5	5	3	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
221	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
222	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
223	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
224	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
225	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
226	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
227	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
228	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
229	5	0.1	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
230	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
231	5	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	5	0.1	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

232	5	5	5	5	5	5	5	5
233	0.8	5	3	5	5	5	5	5
234	0.8	5	5	2	5	5	5	5
235	0.1	2	0	0	0	0	0	2
236	0.8	5	5	5	5	5	5	5
237	0.1	3	3	3	3	3	2	5
238	5	5	5	5	5	5	5	5
	0.8	2	5	4	5	5	5	5
	0.1	5	4	3	5	5	5	5
239	0.8	2	5	5	5	5	5	5
	0.1	5	3	3	5	5	5	5
240	5	5	5	5	5	5	5	5
	0.8	5	4	5	5	5	5	5
	0.1	5	3	5	5	5	5	5
241	0.8	5	3	5	5	5	5	5

- to be cont'd -

242	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	4	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
243	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	4	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
244	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	0	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
245	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	0	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
246	5	5	4	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
247	0.8	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	0	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
248	5	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	4	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	0	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
249	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
250	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	1	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
251	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	0	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

252	5	5	5	5	5	5	5	5
	0.8	3	5	5	5	5	5	5
253	0.8	5	4	5	5	5	5	5
	0.1	4	5	5	5	5	5	5
254	0.8	5	5	5	5	5	5	5
	0.1	4	5	5	5	5	5	5
255	0.2	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5
256	0.2	5	5	5	5	5	5	5
	0.1	3	5	5	5	5	5	5
257	0.2	5	5	5	5	5	5	5
	0.1	3	5	5	5	3	5	5
258	0.2	5	5	5	5	5	5	5
	0.1	4	5	5	5	5	5	5
259	0.8	5	5	5	5	5	5	5
	0.2	4	5	5	5	5	5	5
	0.1	3	4	5	5	5	5	5
260	0.8	5	3	5	5	5	5	5
	0.2	3	4	5	5	5	5	5
261	0.2	5	5	5	5	5	5	5
	0.1	3	3	5	5	5	5	5

- to be cont'd -

262	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
263	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	4
264	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	3	5	5
265	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	4
266	0.2	5	5	5	5	5	5	5	5	5	5	5	4	5	4	3	5	4	3	5	3
267	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
268	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
269	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
270	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

271	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	4
272	0.1	3	5	5	5	5	5	5	5	5	5	5	3	5	4	5	4	2	0	4	2
273	0.2	5	5	5	5	5	5	5	5	5	5	5	3	5	5	5	5	5	5	5	4
	0.1	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
274	0.2	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
	0.1	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
275	0.8	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
	0.2	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
	0.1	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
276	5	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
	0.8	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
277	5	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
	0.8	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
278	5	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
	0.8	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
279	5	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1
	0.8	5	3	5	3	5	4	3	5	4	5	3	2	5	3	5	2	5	2	4	1

- to be cont'd -

280	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
281	5	5	5	4	4	4	5	5	5	5	5	5	3	5	5	5	5	5	5	0
	0.8	5	5	4	5	5	5	5	5	5	5	5	0	5	3	5	5	3	5	5
282	5	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
	0.8	3	5	3	5	3	5	3	5	3	5	4	3	5	4	5	4	3	5	3
283	5	5	5	3	5	3	5	3	5	3	5	4	3	5	4	5	4	3	5	3
	0.8	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
284	5	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	0	5	3	5	5	3	5	5
285	5	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
	0.8	3	5	3	5	3	5	3	5	3	5	4	3	5	4	5	4	3	5	3
	0.1	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
286	5	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	0	5	3	5	5	3	5	5
287	5	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
	0.8	3	5	3	5	3	5	3	5	3	5	4	3	5	4	5	4	3	5	3
	0.1	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
288	5	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	0	5	3	5	5	3	5	5
	0.1	5	5	4	5	5	5	5	5	5	5	5	3	5	4	5	5	5	5	5

- to be cont'd -

289	0.8	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	4	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
290	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	3	5	4	5	5	5	4	0	5	5	0	5	5	0	5	5	5	5	0
291	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	3	5	5	5	5	5	5	2	5	5	2	5	5	3	5	5	5	5	0
292	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	4
	0.8	5	4	5	5	5	5	5	5	2	5	5	2	5	5	3	5	5	5	5	0
293	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	3	5	4	5	5	5	4	0	5	5	0	5	5	0	5	5	5	5	0
294	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	3	5	3	5	3	5	4	3	5	4	3	5	4	2	5	3	5	4	2
295	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	4
	0.1	5	3	5	4	5	5	5	4	0	5	5	0	5	5	0	5	5	5	5	0
296	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	3	5	4	5	5	5	4	0	5	5	0	5	5	0	5	5	5	5	0
	0.1	5	3	5	4	5	5	5	4	0	5	5	0	5	5	0	5	5	5	5	0

- to be cont'd -

297	5	0.8	5	5	5	3	5	3	5
298	5	0.8	5	4	5	3	5	5	3
299	5	0.8	5	4	5	3	5	5	5
300	5	0.8	5	2	5	3	5	5	5
301	5	0.8	5	5	5	3	5	5	5
302	5	0.8	5	4	5	3	5	4	5
		0.8		4		3		4	
		0.1		0		3		4	
303	5		5	5	5	3	5	5	5
		0.8		5		3		5	
304		0.8		3		3		5	
		0.1		0		3		4	
305		0.8		5		3		5	
		0.1		0		3		5	
306	5	0.8	5	4	5	3	5	5	5

- to be cont'd -

307	0.8	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	0	3	2	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
308	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	0	3	2	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
309	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	0	3	2	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
310	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	0	3	2	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
311	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	0	3	2	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
312	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	0	3	2	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
313	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	0	3	2	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3

- to be cont'd -

314	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0.1	5	0.8	0.1	5	0.8	0.1	5	0.8	0.1	5	0.8	0.1	5	0.8	0.1	5	0.8	5	5	5	5
315	5	4	4	5	5	0	5	5	0	5	5	0	5	4	0	5	3	3	5	5	2	5	5
316	5	5	0	5	5	5	5	5	4	5	5	4	5	5	2	5	4	2	5	4	5	5	5
317	5	5	3	5	5	5	5	5	5	5	5	5	5	5	3	5	5	4	5	5	5	5	5
318	5	4	2	5	4	3	5	4	3	5	4	2	5	4	2	5	4	2	5	3	3	3	3
319	5	4	2	5	4	3	5	4	3	5	4	2	5	4	2	5	4	2	5	3	3	3	3
320	5	4	2	5	4	3	5	4	3	5	4	2	5	4	2	5	4	2	5	3	3	3	3
321	5	4	2	5	4	3	5	4	3	5	4	2	5	4	2	5	4	2	5	3	3	3	3

- to be cont'd -

321	0.8	1	5	2	5	5
322	0.8	5	4	5	5	5
	0.1	3	1	3	5	5
323	0.8	5	5	5	5	5
	0.1	5	5	5	5	5
324	0.8	5	5	5	5	5
	0.1	5	5	5	5	5
325	0.8	5	5	5	5	5
	0.1	5	5	5	5	5
326	0.8	5	5	5	5	5
	0.1	5	5	5	5	5
327	0.8	5	5	5	5	5
	0.1	5	5	5	5	5
328	0.8	5	5	5	5	5
	0.1	5	5	5	5	5
329	5	5	5	5	4	5
330	0.8	2	5	3	2	5
	5	3	5	4	2	5
331	0.1	1	3	2	0	4
	0.8	5	5	5	5	5
	0.1	2	3	5	0	5

- to be cont'd -

332	0.8	5	5	5	5	5	5	5	5	5
333	0.1	3	5	0	5	5	5	5	5	5
334	5	5	5	5	5	5	5	5	5	5
335	0.8	2	5	5	5	5	5	5	5	5
336	5	0.8	2	5	5	5	5	5	5	5
337	5	0.8	4	5	5	5	5	5	5	5
338	5	0.8	1	5	5	5	5	5	5	5
339	5	0.8	5	5	5	5	5	5	5	5
340	5	0.8	3	5	5	5	5	5	5	5
341	0.8	0.8	5	5	5	5	5	5	5	5
342	0.2	0.2	2	5	5	5	5	5	4	5

- to be cont'd -

342	0.1	3	5	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
343	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
344	0.1	5	5	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
345	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
346	0.1	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
347	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
348	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
349	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
350	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
351	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

352	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
353	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
354	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
355	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
356	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
357	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
358	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
359	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
360	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
361	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

362	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
363	0.1	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
364	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
365	0.1	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
366	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
367	0.1	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
368	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
369	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
370	0.1	3	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
371	0.2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

- to be cont'd -

372	0.2	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	4	5
373	0.8	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5
374	0.8	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5
375	0.8	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5
376	0.8	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	5	5
377	0.2	5	5	5	5	5	5	5	5
	0.1	5	5	5	5	5	5	0	5
378	5	5	5	5	5	5	5	5	5
	0.8	2	2	5	5	5	5	4	5
379	5	5	5	5	5	5	5	3	5
	0.8	4	4	4	4	4	4	-	5
380	5	5	5	5	5	5	5	5	5
	0.8	2	5	5	5	5	5	4	5
381	5	5	5	5	5	5	5	5	5
	0.8	3	4	5	5	5	5	5	5

- to be cont'd -

382	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
383	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
384	5	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
385	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
386	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
387	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
388	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
389	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
390	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

391	5	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
392	5	0.8	5	4	3	2	0	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5
393	5	0.8	5	5	4	2	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
394	5	0.8	5	5	5	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
395	5	0.8	5	5	5	5	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	5	0.8	5	5	5	5	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	5	0.1	1	4	1	4	0	5	5	5	5	5	4	4	4	4	4	4	4	4	4	4	4
396	5	0.8	3	5	3	5	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
397	5	0.8	5	5	5	5	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
398	5	0.8	5	5	5	5	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
399	5	0.8	5	5	5	5	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
400	5	0.8	5	5	5	5	0	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

401	5	5	5	5	5	5	0
	0.8	5	5	5	5	5	0
402	5	5	5	5	5	5	0
	0.8	4	5	5	5	5	0
403	0.2	5	5	5	4	5	0
	0.1	4	3	2	0	3	0
404	0.2	5	5	5	5	5	0
	0.1	5	5	5	5	5	0
405	0.2	5	5	5	4	5	0
	0.1	3	3	3	3	4	0
406	0.2	4	4	4	3	5	0
	0.1	3	3	3	0	3	0
A	5	0	0	0	0	0	0
B	5	0	0	0	0	2	0
C	5	0	0	0	0	0	0
D	5	0	0	0	0	0	0

As shown in table 6, the present 3-(substituted phenyl)pyrazole derivatives or salt thereof, represented by the general formula (I), exhibit more excellent controlling effects of weeds than comparative compound A, B, C or D at post-emergence treatment in paddy fields. Even when the derivatives exhibit both herbicidal activity on weeds and phytotoxicity on crops at the same dosage, however, by the selection of a proper lower dosage, sufficient herbicidal activity is retained but phytotoxicity on crops is reduced remarkably.

Test Example 3 Herbicidal effect on upland field weeds of pre-emergence stage.

Polyethylene vats of 10 cm W x 20 cm L x 5 cm depth were filled with soil, and planted separately with seeds of barnyardgrass, velvetleaf, cocklebur, jimsonweed, birdseye speedwell, cleavers, which are upland field weeds, and separately with seeds of soybean and wheat as upland field crops.

Soil in each pot was sprayed with each of solutions containing compounds (listed in Table 1) of the present invention as active ingredient at a predetermined concentration. 14 Days later, the herbicidal effect was examined, the percentages of killed weeds were calculated in the same manner as in Test Example 1, and the phytotoxicity of soybean and wheat plants were also examined and judged according the criterion shown in Test Example 1.

Results of the test are shown in Table 7.

Table 7

Comp'd No.	Amount of active ingredient Kg/ha	Phytotoxicity		Pre-emergence treatment					
		wheat	soybean	banyard-grass	velvet-leaf	cocklebur	jimsonweed	birdseye speedwell	cleavers
3	5	1	0	5	5	5	5	-	5
4	5	2	1	5	5	0	4	-	5
5	5	3	0	5	4	2	5	-	5
6	5	1	2	5	3	0	2	-	3
7	5	0	1	4	4	0	2	-	2
8	5	0	0	3	3	0	1	-	1
9	5	4	4	5	5	-	5	-	5
	0.8	2	0	5	4	3	5	-	-
10	5	5	4	5	5	5	5	-	5
	0.8	3	2	3	5	5	5	-	-
11	5	5	5	5	4	4	5	-	5
12	5	4	4	5	4	-	5	-	5
	0.8	3	1	5	2	-	5	-	-
13	5	4	4	5	5	5	5	-	5
	0.8	3	1	5	5	5	5	-	-

- to be cont'd -

14	5	5	3	5	5	-	5	-	5
	0.8	3	0	5	5	-	5	-	-
16	5	1	-	4	3	2	4	-	5
17	5	0	0	1	0	0	4	-	0
18	5	1	2	4	4	2	4	-	5
19	5	1	1	5	-	1	4	-	4
20	5	1	0	3	0	0	3	-	5
21	5	0	0	4	2	0	1	-	0
23	5	0	0	4	0	0	1	-	0
24	5	0	1	0	0	0	4	-	4
25	5	1	0	3	2	0	5	-	4
26	5	4	3	5	5	-	5	-	5
	0.8	2	1	4	4	0	4	-	4
27	5	4	4	5	5	-	5	-	5
	0.8	2	2	4	4	-	5	-	4
28	5	4	2	4	5	-	5	-	5
	0.8	2	0	3	4	0	5	-	5
29	5	2	3	5	-	2	5	-	4
30	5	3	3	4	5	-	5	-	5
	0.8	1	1	3	4	1	5	-	3

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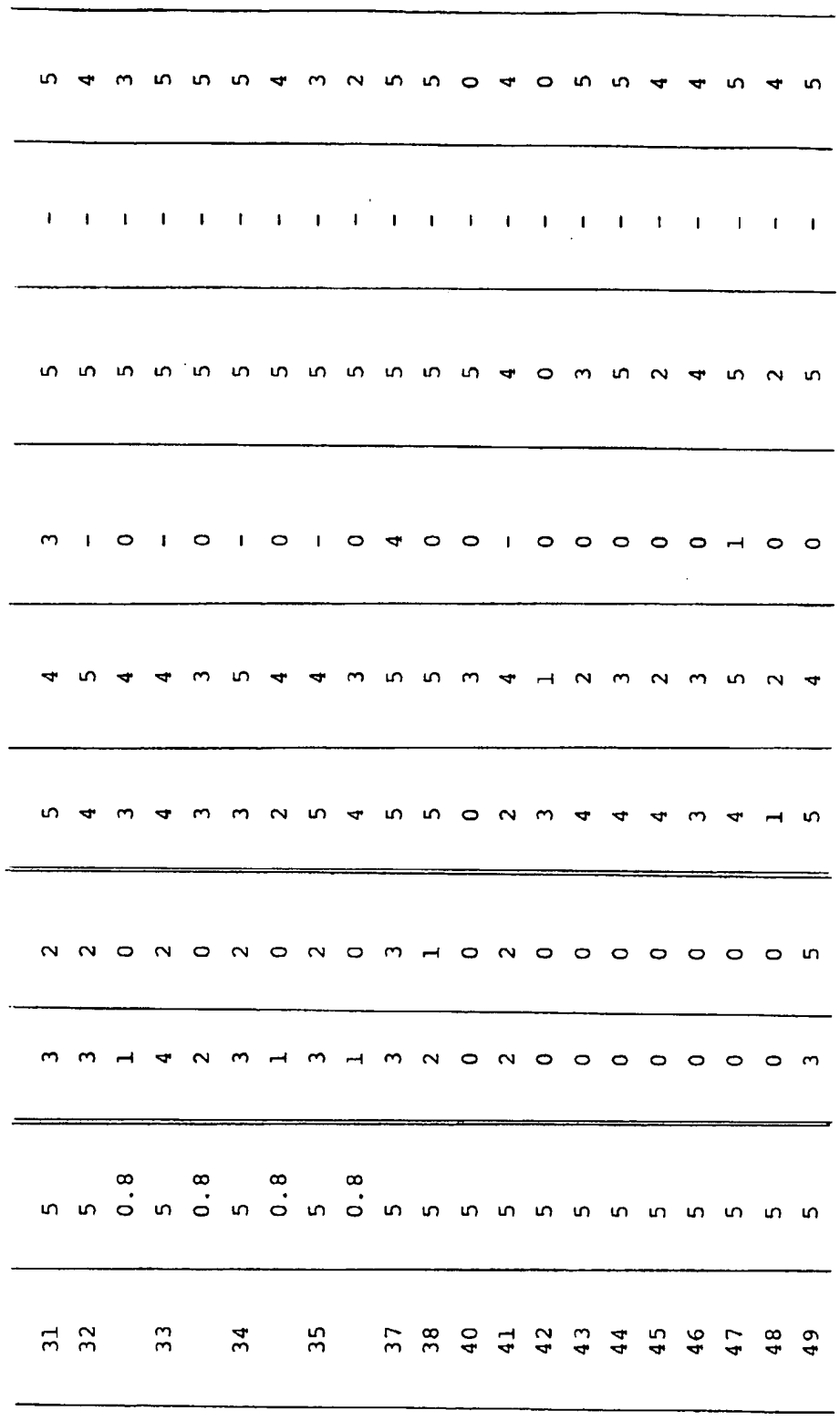

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34	5	5	-	3	5	2	3	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
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	0.8	0	0	2	0	0	0	4	3	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
37	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
38	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
40	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
41	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
42	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
43	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
44	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
45	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
46	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4
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49	5	5	-	5	5	2	4	5	4	-	5	5	0	-	5	4	4	3	4	4	4	3	4	4

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52	5	5	3	5	0	0	0	5	1	1	2	1	1	0	4	2	4	2	3	2
54	5	5	5	5	3	3	4	5	5	4	5	5	5	5	5	5	5	5	5	5
55	4	5	5	5	1	2	1	5	4	5	5	5	5	4	5	5	5	5	5	4
57	-	5	-	5	0	0	0	-	4	0	2	1	-	0	5	4	5	4	-	4
59	5	5	5	5	2	0	2	5	5	4	5	5	3	2	5	5	5	5	5	5
71	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
72	5	5	5	5	1	0	0	5	5	4	5	2	4	2	5	-	5	-	5	5
73	5	5	5	5	2	0	0	5	5	4	5	2	4	2	5	-	5	-	5	5
74	5	5	5	5	1	0	0	5	5	4	5	2	4	2	5	-	5	-	5	5
76	5	5	5	5	1	0	0	5	5	4	5	2	4	2	5	-	5	-	5	5
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82	5	5	5	5	1	0	0	5	5	4	5	2	4	2	5	-	5	-	5	5
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86	5	0.8	5	3	5	2	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
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91	5	0.8	5	4	2	4	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5
92	5	0.8	5	4	2	2	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5
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96	5	0.8	1	1	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
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105	5	4	3	5	5	-	5	-	5
	0.8	3	1	5	5	-	5	-	5
106	5	4	3	5	5	2	-	5	5
	0.8	3	1	5	5	-	-	5	4
107	5	5	3	5	5	3	-	5	5
	0.8	3	1	5	4	-	-	5	5
108	5	4	3	5	5	3	-	5	5
	0.8	3	1	5	5	-	-	5	5
109	5	4	3	5	5	2	5	-	5
	0.8	3	1	5	5	-	5	-	5
110	5	5	3	5	5	2	5	-	5
	0.8	3	1	5	5	-	5	-	5
111	5	4	2	2	5	1	4	-	5
	0.8	1	0	1	4	-	2	-	3
112	5	4	4	5	5	5	5	-	5
	0.8	3	2	5	5	3	5	-	-
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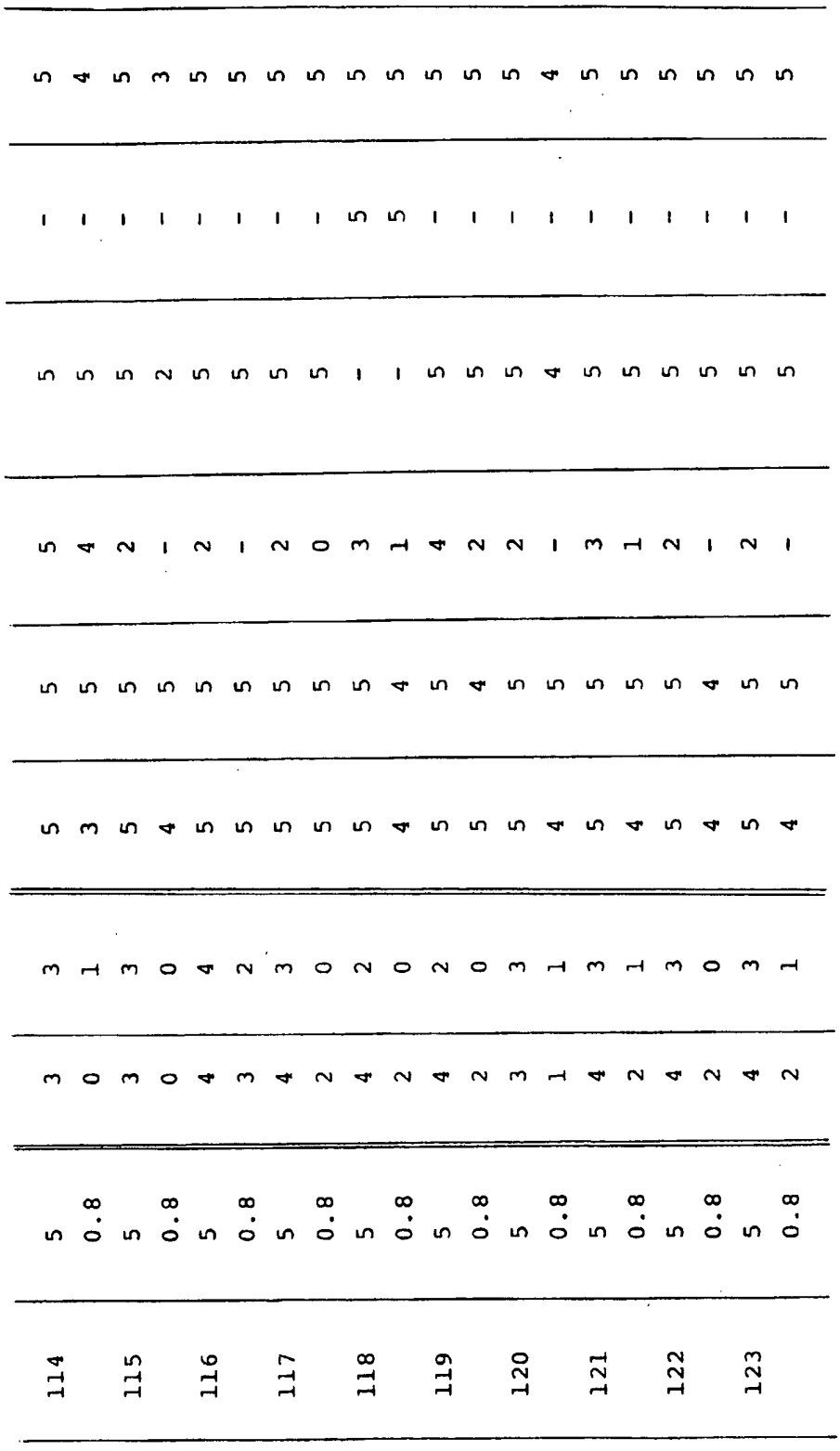

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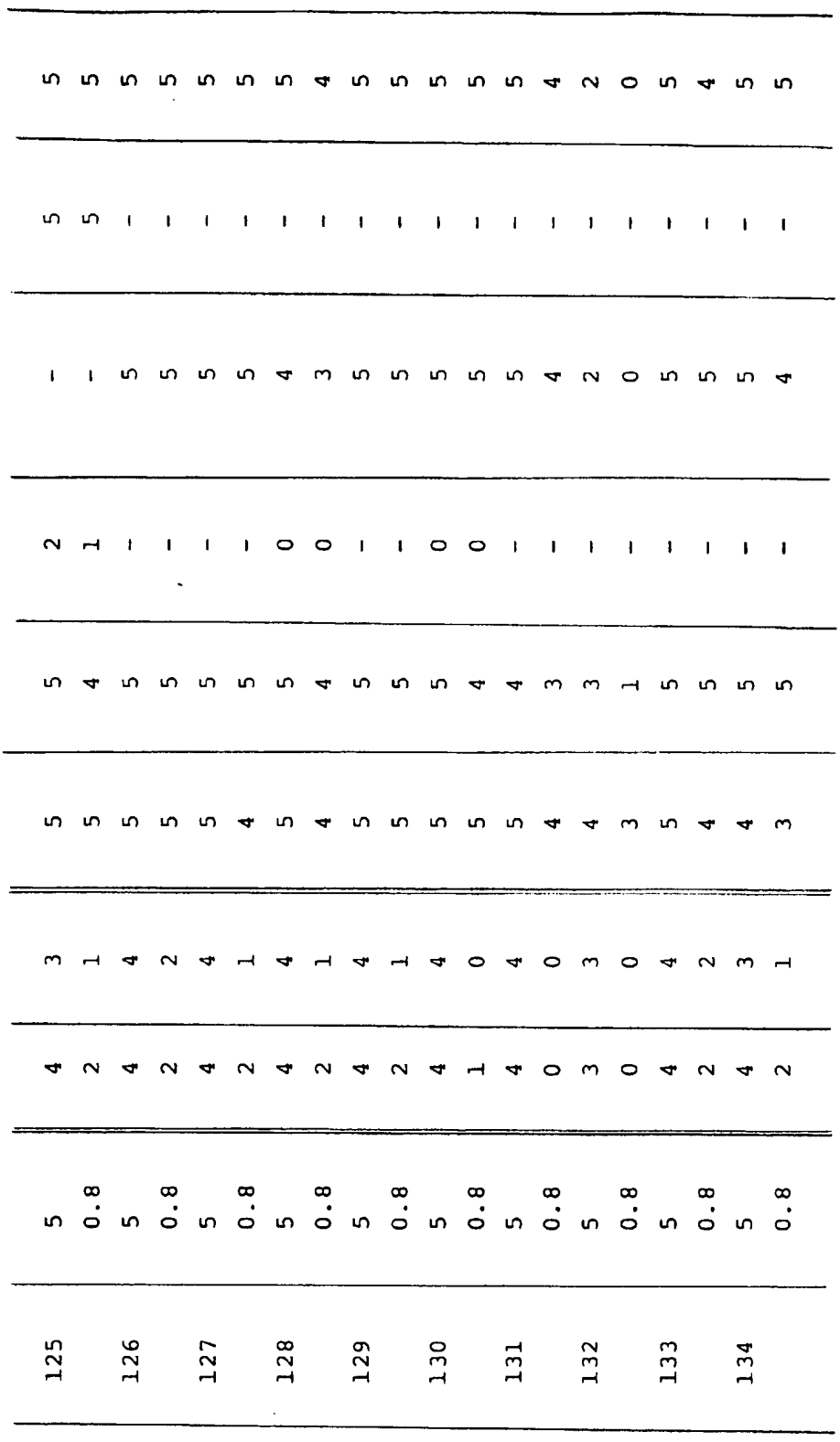

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127	5	0.8	4	2	4	4	5	5	5	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
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133	5	0.8	4	2	4	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
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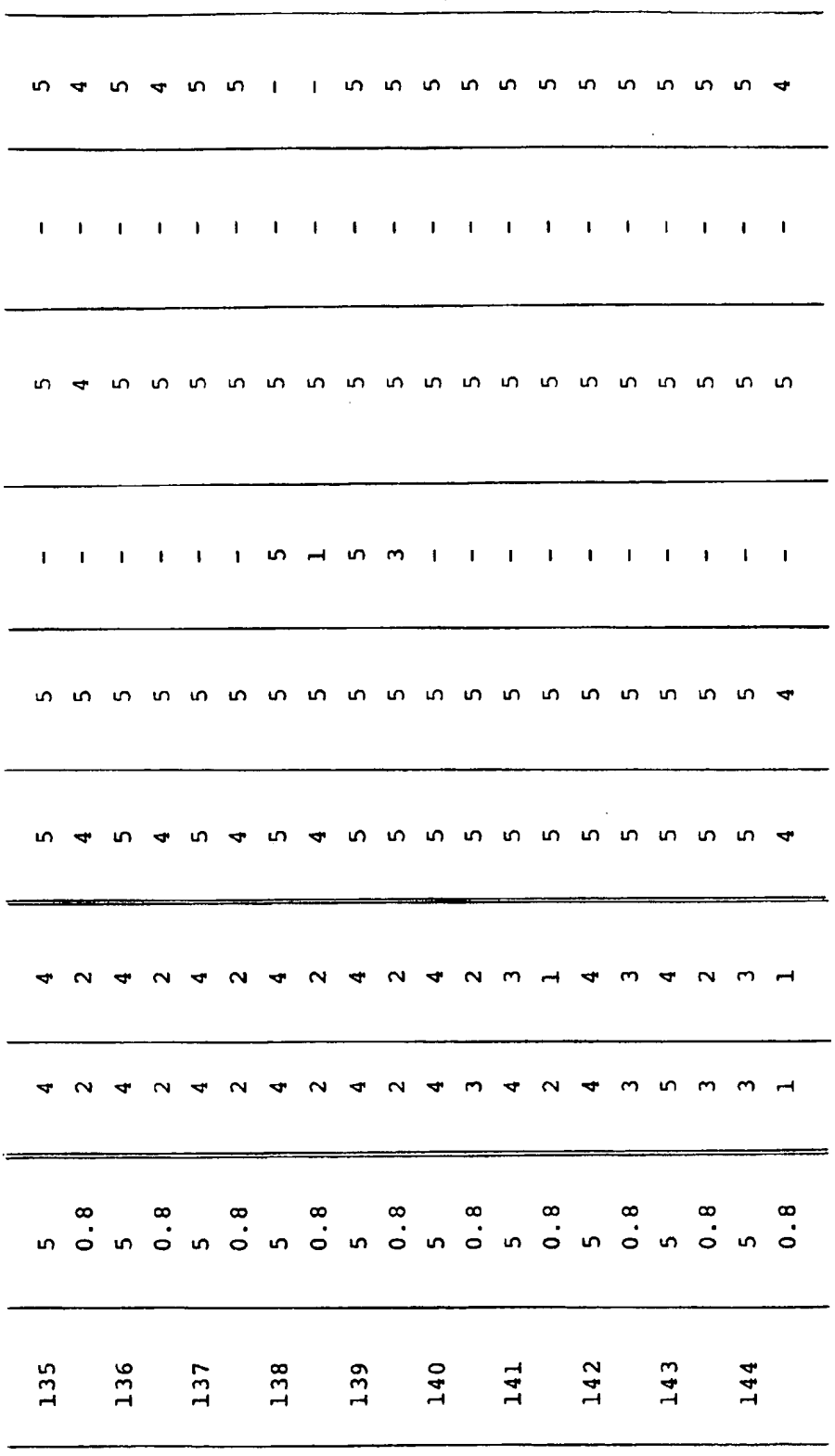

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137	5	0.8	4	2	4	2	4	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	4
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143	5	0.8	3	2	4	2	4	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	4
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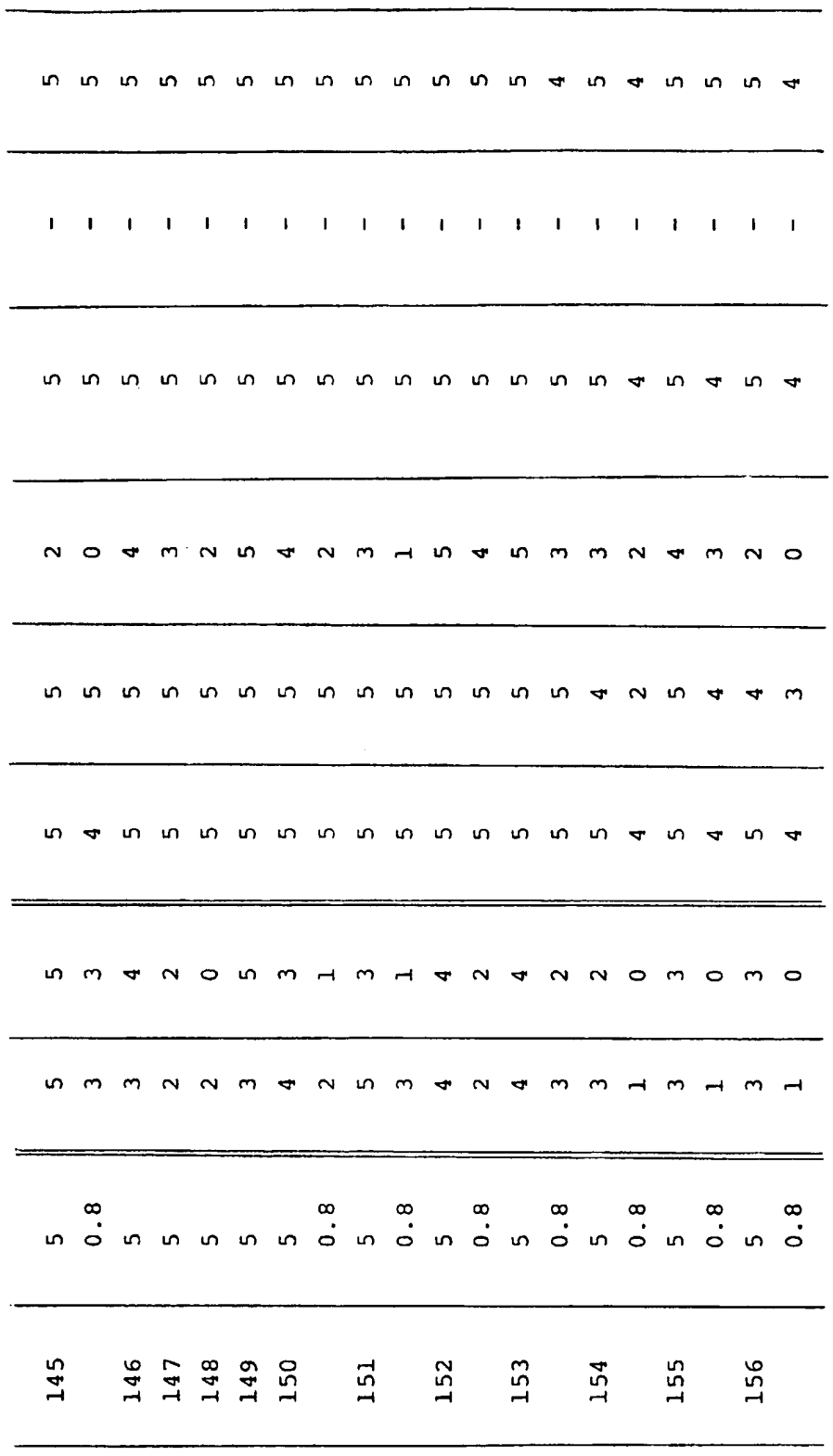

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159	5	0.8	3	0	3	3	4	5	4	5	4	5	4	0	0	0	0	0	0	0	-	-	2
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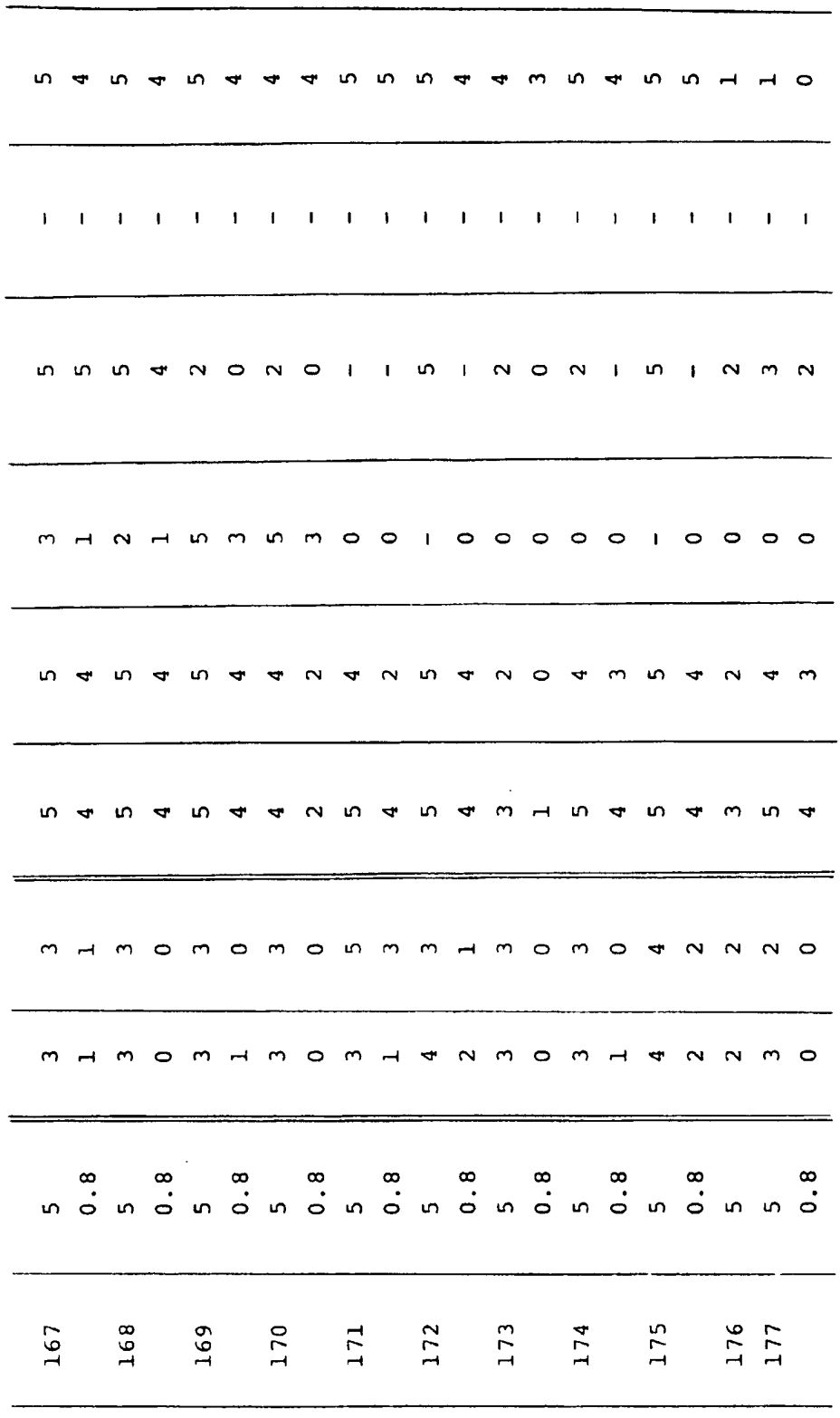

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180	5	3	2	2	4	-	3	-	4
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190	5	3	3	4	4	-	-	5	5
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	0.8	0	0	2	0	0	-	5	0
204	5	3	3	3	4	2	-	5	5
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	0.8	1	1	3	0	0	2	-	4
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212	5	2	1	4	3	0	0	-	5
216	5	3	2	4	3	5	2	-	5
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	0.2	0	0	2	4	0	-	5	1
237	5	3	2	3	5	1	-	5	5
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238	5	4	4	5	5	2	-	5	5
	0.8	2	2	4	5	-	-	5	5
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	0.8	1	1	4	5	-	-	5	5
241	5	3	2	5	5	1	-	5	5
	0.8	1	0	3	4	0	-	5	3
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	0.8	1	0	4	5	0	-	5	3
243	0.8	2	1	4	5	1	-	5	4
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249	5	3	3	5	5	2	-	5	5
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253	5	4	3	5	5	3	-	5	5
	0.8	2	1	4	5	2	-	5	4
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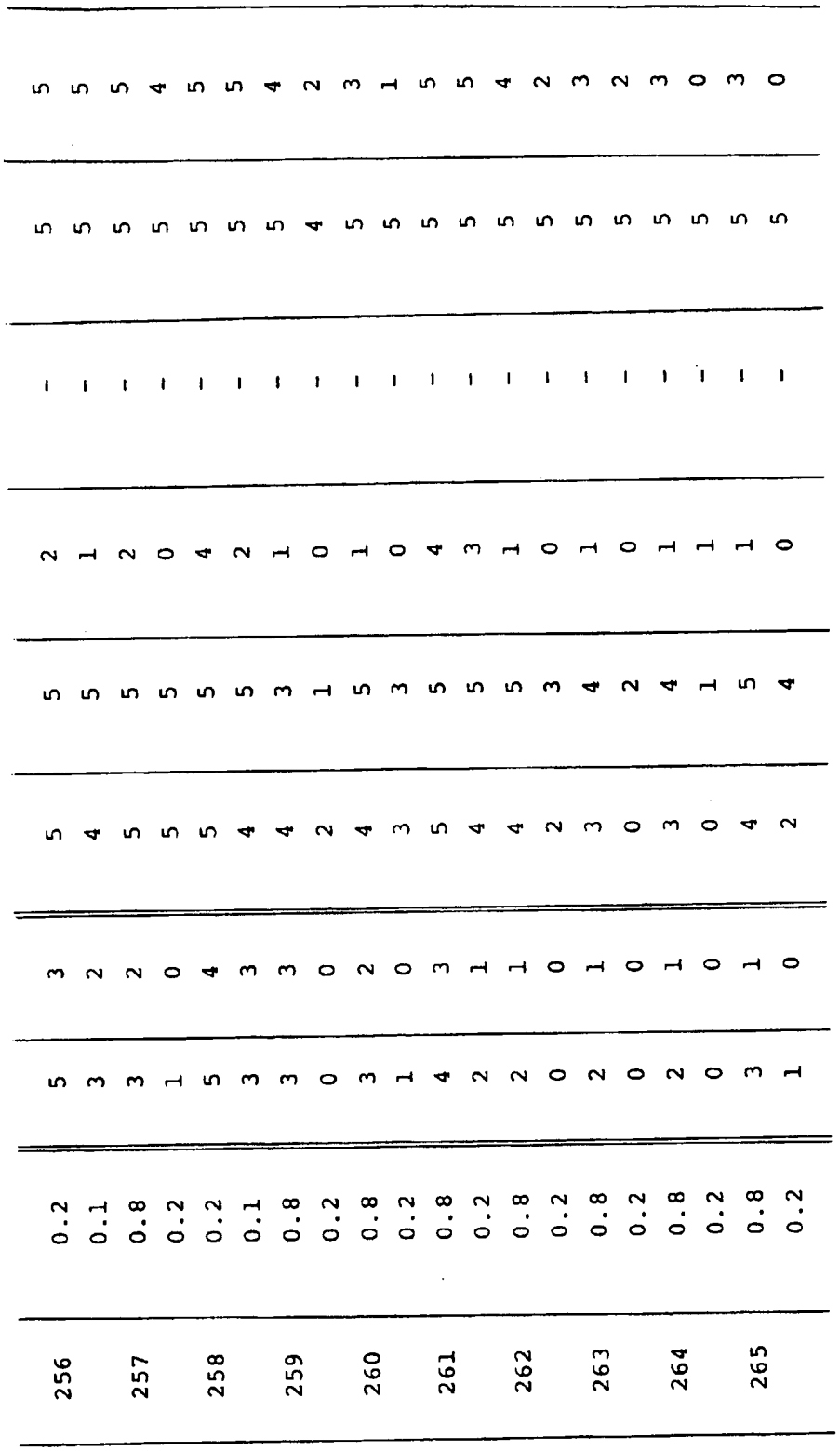

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262	0.2	2	1	3	4	5	4	4	5	5	3	-	5	5	5	5	5	5	5	5	5	5	5
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269	0.8	3	1	5	4	1	0	5	-	5	4
	0.2	1	0	2	2	0	0	5	-	5	2
270	0.8	3	1	5	5	1	1	5	-	5	4
	0.2	1	0	2	5	1	0	5	-	5	2
271	0.8	3	1	5	5	1	0	5	-	5	5
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272	0.8	3	1	5	5	2	0	5	-	5	5
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275	0.8	3	1	5	5	1	1	5	-	5	5
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- to be cont'd -

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278	5	3	3	4	5	1	-	5	4
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279	5	3	3	4	5	1	-	5	4
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280	5	4	2	5	5	1	-	5	5
	0.8	2	0	4	3	0	-	5	5
281	5	3	3	5	5	1	-	5	5
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282	5	3	3	5	5	2	-	5	5
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283	5	3	3	5	5	2	-	5	5
	0.8	1	1	4	5	0	-	5	5
284	5	3	3	5	5	1	-	5	5
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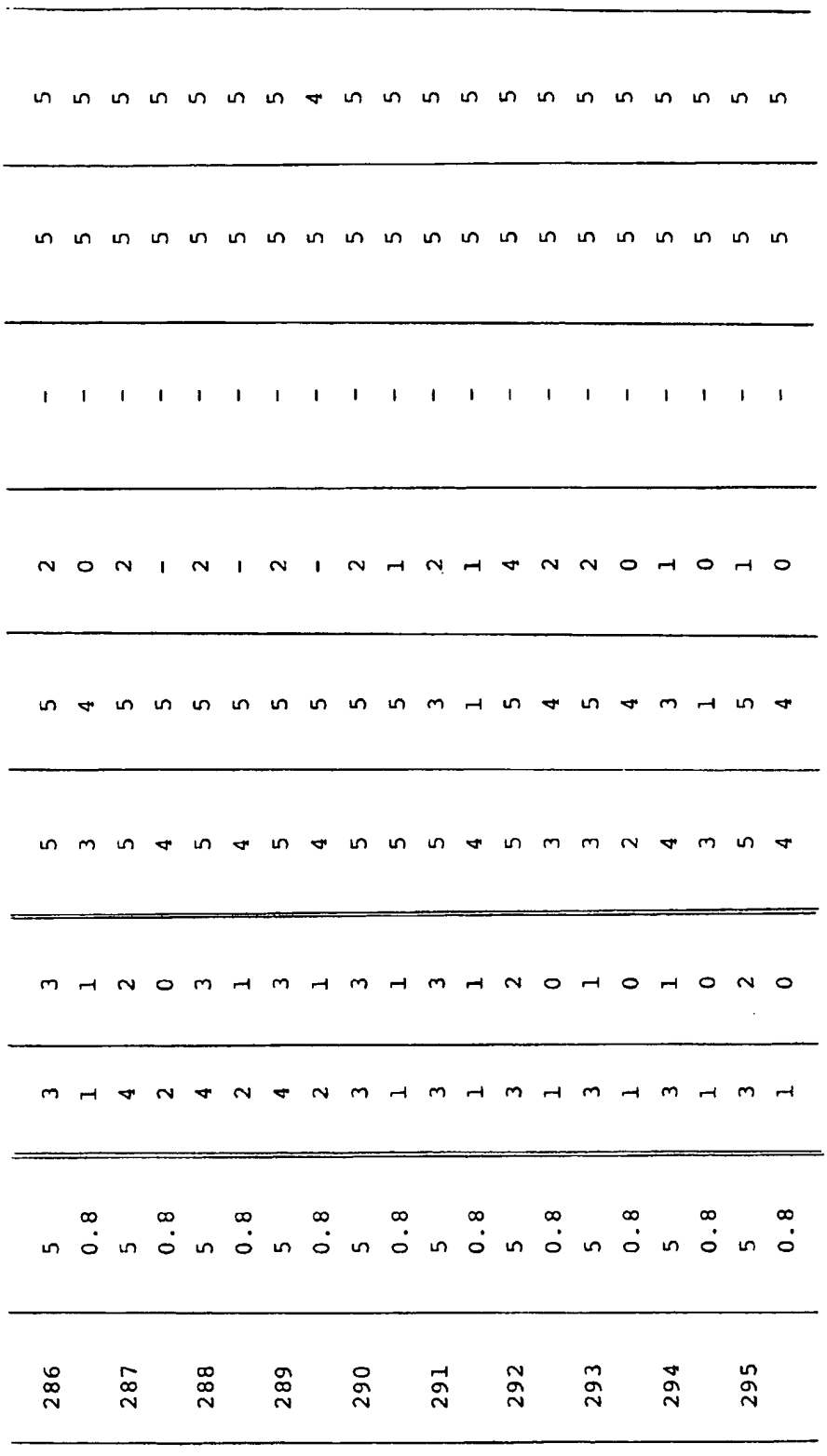

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298	5	3	3	4	5	1	-	5	5
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299	5	4	3	5	5	1	-	5	5
	0.8	2	1	3	4	0	-	5	4
300	5	3	2	4	5	1	-	5	5
	0.8	1	0	2	3	0	-	5	4
302	5	4	2	5	5	2	-	5	5
	0.8	2	0	4	5	0	-	5	5
301	5	3	2	4	5	1	-	5	5
	0.8	1	0	3	3	0	-	5	5
303	5	4	2	4	5	1	-	5	5
	0.8	2	0	3	3	0	-	5	5
304	5	3	3	5	5	2	-	5	5
	0.8	1	1	4	4	1	-	5	5
305	5	3	3	5	5	1	-	5	5
	0.8	1	1	4	5	0	-	5	3

- to be cont'd -

306	5	4	3	4	5	2	-	5	5
	0.8	2	1	3	3	1	-	5	5
307	5	3	2	5	5	2	-	5	5
	0.8	1	0	4	4	0	-	5	5
308	5	4	3	5	5	2	-	5	5
	0.8	2	1	4	5	-	-	5	4
309	5	4	4	5	5	2	-	5	5
	0.8	2	2	4	5	-	-	5	5
310	5	4	5	5	5	2	-	5	5
	0.8	2	3	3	5	0	-	5	5
312	5	3	2	3	4	1	-	5	5
	0.8	1	0	2	2	0	-	5	5
313	5	3	3	5	5	2	-	5	5
	0.8	1	1	4	5	0	-	5	5
314	5	3	2	4	5	2	-	5	5
	0.8	1	0	2	5	0	-	5	5
315	5	3	3	5	5	1	-	5	5
	0.8	1	1	3	4	0	-	5	5
316	5	4	3	5	5	2	-	5	5
	0.8	2	1	3	4	1	-	5	5

- to be cont'd -

317	5	0.8	4	3	5	3	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5
318	5	0.8	2	1	4	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
319	5	0.8	3	2	3	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
320	5	0.8	1	0	4	1	1	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
321	5	0.8	3	3	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
322	5	0.8	1	0	2	2	3	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
323	5	0.8	2	0	1	1	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
324	5	0.8	5	4	2	0	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
325	5	0.8	3	2	4	2	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
326	5	0.8	4	2	3	1	4	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

327	5	4	3	5	5	5	-	5	5
	0.8	2	1	4	5	2	-	5	5
328	5	3	4	5	5	4	-	5	5
	0.8	1	2	4	5	2	-	5	4
329	5	2	1	3	3	1	-	5	4
	0.8	0	0	1	0	0	-	5	1
330	5	3	1	4	5	1	-	5	5
331	5	5	5	5	5	5	-	5	5
	0.8	3	4	5	5	5	-	5	5
332	0.8	5	5	5	5	5	-	5	5
	0.2	3	3	3	4	3	-	5	4
333	5	3	2	4	5	1	-	5	2
	0.8	1	0	3	3	1	-	5	-
334	5	2	2	3	4	1	-	5	5
	0.8	0	0	2	2	0	-	5	5
335	5	2	1	4	5	1	-	5	5
	0.8	0	0	0	0	0	-	4	-
336	5	2	2	5	5	2	-	5	5
	0.8	0	0	4	5	1	-	5	4
337	5	2	2	4	3	1	-	5	5
	0.8	0	0	3	1	0	-	5	3

- to be cont'd -

338	5	3	2	4	5	1	-	5	5
	0.8	1	0	3	3	0	-	5	5
339	5	2	1	4	4	0	-	5	4
340	5	2	2	3	4	1	-	5	5
	0.8	0	0	2	2	0	-	5	5
341	0.8	3	1	3	4	1	-	5	4
	0.2	1	0	0	1	0	-	3	3
342	0.8	3	1	5	5	1	-	5	4
	0.2	1	0	4	5	0	-	5	2
343	0.8	3	1	5	5	1	-	5	4
	0.2	1	0	4	5	0	-	5	2
344	0.8	3	1	5	5	1	-	5	4
	0.2	1	0	3	3	0	-	5	2
345	0.8	3	2	5	5	1	-	5	4
	0.2	1	1	3	4	0	-	5	2
346	0.8	3	1	5	5	1	-	5	4
	0.2	1	0	3	4	0	-	5	2
347	0.8	3	1	5	5	1	-	5	4
	0.2	1	0	3	4	0	-	5	1
348	0.8	3	1	5	5	1	-	5	4
	0.2	1	0	3	5	0	-	5	1

- to be cont'd -

349	0.8	3	1	4	2	5	1	-	5	4	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	1	0	2	0	4	0	-	5	4	0	0	0	0	0	0	0	0	0	0	0	0	0
350	0.8	3	1	4	2	5	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	1	0	2	0	3	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
351	0.8	2	1	3	1	4	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	0	0	0	0	3	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
352	0.8	3	1	4	2	5	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	1	0	2	0	4	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
353	0.8	2	1	4	2	5	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	0	0	2	0	4	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
354	0.8	3	1	4	2	4	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	1	0	2	0	2	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
355	0.8	2	1	4	2	4	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	0	0	2	0	2	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
356	0.8	2	1	4	2	4	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	1	0	1	0	1	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
357	0.8	2	1	4	2	4	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	0	0	3	0	1	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
358	0.8	2	1	4	2	5	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0
	0.2	0	0	3	0	4	1	-	5	5	0	0	0	0	0	0	0	0	0	0	0	0	0

- to be cont'd -

359	0.8	3	1	1	5	3	5	5	5	-	1	5	5	4	1	5	3	5	5	5	5	4
360	0.2	1	0	0	5	5	5	5	5	-	0	5	5	5	0	5	3	5	5	5	5	1
361	0.8	3	1	1	5	5	5	5	5	-	1	5	5	5	1	5	4	5	5	5	5	5
362	0.2	1	0	0	5	5	5	5	5	-	0	5	5	5	0	5	3	5	5	5	5	0
363	0.8	3	1	1	5	5	5	5	5	-	1	5	5	5	1	5	4	5	5	5	5	3
364	0.8	3	1	1	5	5	5	5	5	-	0	5	5	5	1	5	4	5	5	5	5	4
365	0.2	1	0	0	5	5	5	5	5	-	1	5	5	5	0	5	3	5	5	5	5	5
366	0.8	3	1	1	5	5	5	5	5	-	0	5	5	5	1	5	4	5	5	5	5	4
367	0.2	0	0	0	5	5	5	5	5	-	1	5	5	5	0	5	3	5	5	5	5	5
368	0.8	2	1	1	5	5	5	5	5	-	1	5	5	5	1	5	4	5	5	5	5	5

- to be cont'd -

369	0.8	3	2	5	5	2	-	5	5	5
	0.2	1	2	4	5	0	-	5	5	5
370	0.8	3	2	4	5	2	-	5	5	5
	0.2	0	1	2	4	0	-	4	4	4
371	0.8	3	1	5	5	1	-	5	5	5
	0.2	1	0	3	4	0	-	5	5	5
372	0.8	4	4	5	5	5	-	5	5	5
	0.2	2	2	5	4	4	-	5	5	5
373	0.8	4	4	5	5	4	-	5	5	5
	0.2	2	2	5	5	3	-	5	5	5
374	0.8	5	4	5	5	4	-	5	5	5
	0.2	3	2	5	5	2	-	5	5	5
375	0.8	5	4	5	5	4	-	5	5	5
	0.2	3	2	5	5	2	-	5	5	5
376	0.8	5	4	5	5	4	-	5	5	5
	0.2	3	2	5	5	2	-	5	5	5
377	0.8	3	3	5	5	5	-	5	5	5
	0.2	1	1	4	5	3	-	5	5	4
378	5	3	1	5	5	1	-	5	5	5
	0.8	0	0	1	0	0	-	4	4	-

- to be cont'd -

379	5	0.8	3	1	4	5	3	1	2	1	5	5	5	5	5	5	5	5	5	5	5
380	5	0.8	3	1	3	3	1	1	2	1	5	5	5	5	5	5	5	5	5	5	5
381	5	0.8	2	0	2	2	0	1	2	1	5	5	5	5	5	5	5	5	5	5	5
382	5	0.8	2	0	3	5	2	1	2	1	5	5	5	5	5	5	5	5	5	5	5
383	5	0.8	3	1	4	5	3	1	2	1	5	5	5	5	5	5	5	5	5	5	5
384	5	0.8	3	1	4	4	4	1	2	1	5	5	5	5	5	5	5	5	5	5	5
385	5	0.8	4	1	3	2	4	1	2	1	5	5	5	5	5	5	5	5	5	5	5
386	5	0.8	3	1	4	5	3	1	2	1	5	5	5	5	5	5	5	5	5	5	5
388	5	0.8	3	0	2	3	5	1	2	1	5	5	5	5	5	5	5	5	5	5	5
389	5	0.8	1	0	4	4	5	1	2	1	5	5	5	5	5	5	5	5	5	5	5

- to be cont'd -

390	5	3	3	4	5	1	-	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	1	1	3	5	0	-	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
391	5	2	3	4	3	0	-	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
	0.8	0	1	3	4	0	-	5	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
392	5	1	2	4	4	1	-	5	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
	0.8	0	0	2	4	1	-	5	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
393	5	1	2	4	3	0	-	5	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
	0.8	0	0	2	4	1	-	5	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
394	5	1	3	4	3	0	-	5	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
	0.8	0	0	2	4	1	-	5	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
395	5	1	3	4	3	0	-	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0	1	3	4	0	-	5	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
396	5	1	2	4	4	1	-	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
	0.8	0	0	2	4	1	-	5	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
398	5	1	3	4	3	0	-	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
	0.8	0	0	2	4	1	-	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
399	5	1	2	4	4	1	-	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
	0.8	0	0	2	4	1	-	5	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
400	5	1	2	4	4	1	-	5	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
	0.8	0	0	2	4	1	-	5	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
401	5	1	3	4	3	0	-	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.8	0	1	3	4	0	-	5	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3

- to be cont'd -

402	5	3	3	5	5	5	-	5	5
	0.8	1	1	4	5	4	-	5	5
403	0.8	4	3	5	5	2	-	5	5
	0.2	2	1	4	5	1	-	5	4
404	4	5	4	5	5	3	-	5	5
	0.8	3	2	5	5	1	-	5	5
405	5	4	3	4	5	1	-	5	5
	0.8	2	1	3	4	0	-	5	4
406	5	4	3	4	5	1	-	5	5
	0.8	2	1	3	4	0	-	5	4
A	5	0	0	0	0	0	0	0	0
B	5	0	0	0	0	0	0	0	0
C	5	0	0	0	0	0	0	0	0
D	5	0	0	0	0	0	0	0	0

As shown in table 7, the present 3-(substituted phenyl)-pyrazole derivatives or salt thereof, represented by the general formula (I), exhibit more excellent controlling effects of weeds than comparative compound A, B, C or D at pre-emergence treatment in paddy fields. Ever when the derivatives exhibit both herbicidal activity on weeds and phytotoxicity on crops at the same dosage, however, by the selection of a proper lower dosage, sufficient herbicidal activity is retained but phytotoxicity on crops is reduced remarkably.

Test Example 4 Herbicidal effect on upland field weeds of post-emergence stage.

Polyethylene vats of 10 cm W x 20 cm L x 5 cm depth were filled with soil, and planted with seeds of upland field weeds shown below and with seeds of soybean and wheat as upland field crops. These weeds and crops were grown to leaf stages shown below. Then these weeds and crops were sprayed separately with solutions containing compounds (listed in Table 1) of the present invention as active ingredient at a predetermined concentration. 14 Days later, the herbicidal effect was examined, the percentages of killed weeds were calculated in the same manner as in Test Example 1, and the phytotoxicity of soybean and wheat plants were examined and judged according to the criterion shown in Test Example 1.

Species of test weeds and leaf stages thereof and leaf stages of test soybean and wheat plants

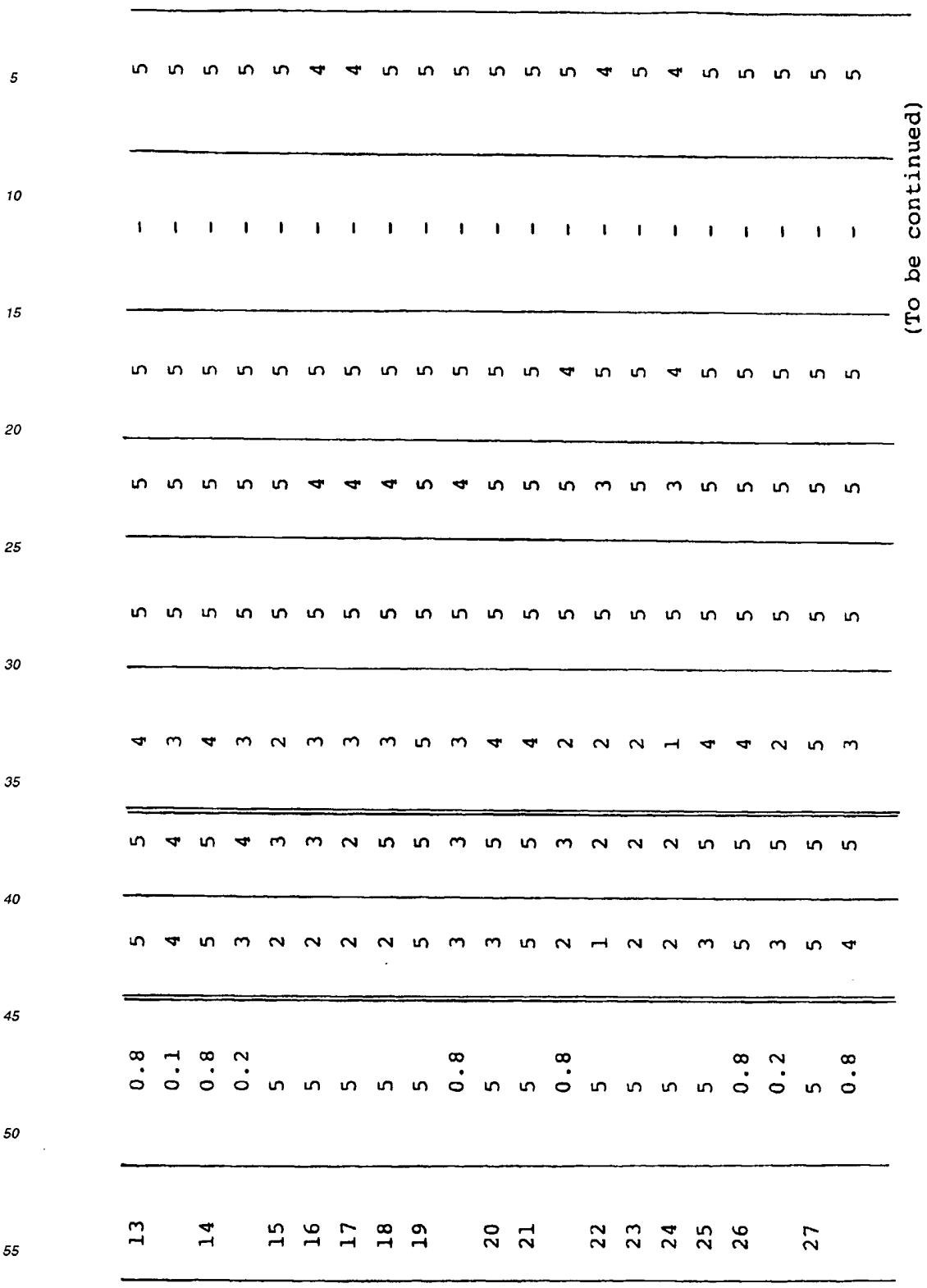
Barnyardgrass	2-leaf stage
Velvetleaf	2 "
Cocklelur	1 "
Jimsonweed	1 "
Birdseye Speedwell	1 "
Cleavers	2 "
Wheat	2 "
Soybean	1 "

Results of the test are shown in Table 8.

Table 8

Com'd. No.	Amount of active ingredient Kg/ha	Phyto-toxicity		post-emergence treatment					
		wheat	soy-bean	banyard-grass	velvet-leaf	cock-lebur	jimson-weed	birdseye speedwell	cleavers
2	5	1	2	2	5	4	5	-	-
3	5	2	4	5	4	4	4	-	4
4	5	5	5	5	5	5	5	-	5
	0.2	3	4	4	5	5	5	-	5
5	5	5	5	5	5	5	5	-	5
	0.2	3	4	5	5	5	5	-	5
6	5	3	5	5	5	5	5	-	5
7	5	2	4	4	5	5	5	-	4
8	5	1	4	2	5	2	5	-	1
9	5	5	5	4	5	5	5	-	5
	0.8	3	5	2	5	5	5	-	5
10	0.8	5	5	5	5	5	5	-	5
	0.1	4	5	4	5	5	5	-	5
11	5	5	5	5	5	5	5	-	5
	0.1	4	4	4	5	5	5	-	5
12	0.8	5	5	4	5	5	5	-	5
	0.2	3	4	3	5	5	5	-	5

(To be continued)



13	0.8	5	4	5	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.1	4	3	4	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
14	0.8	5	4	5	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	3	3	4	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
15	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
16	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
17	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
18	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
19	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
	0.8	5	4	5	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
20	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
21	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
	0.8	5	4	5	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
22	5	1	1	2	3	3	3	3	3	3	2	3	3	3	3	3	3	3	3	3	3	3	3	3
23	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
24	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
25	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
26	0.8	5	5	5	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
	0.2	3	3	4	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5
27	5	2	2	3	4	4	4	4	4	4	3	4	4	4	4	4	4	4	4	4	4	4	4	4
	0.8	4	4	5	5	5	5	5	5	5	4	5	5	5	5	5	5	5	5	5	5	5	5	5

(To be continued)

28	0.8	5	5	4	5	5	5	-	5
	0.1	3	4	2	5	5	5	-	5
29	5	5	5	5	5	5	5	-	5
	0.1	4	5	4	5	5	5	-	5
30	5	5	5	5	5	5	5	-	5
	0.8	4	5	3	5	5	5	-	5
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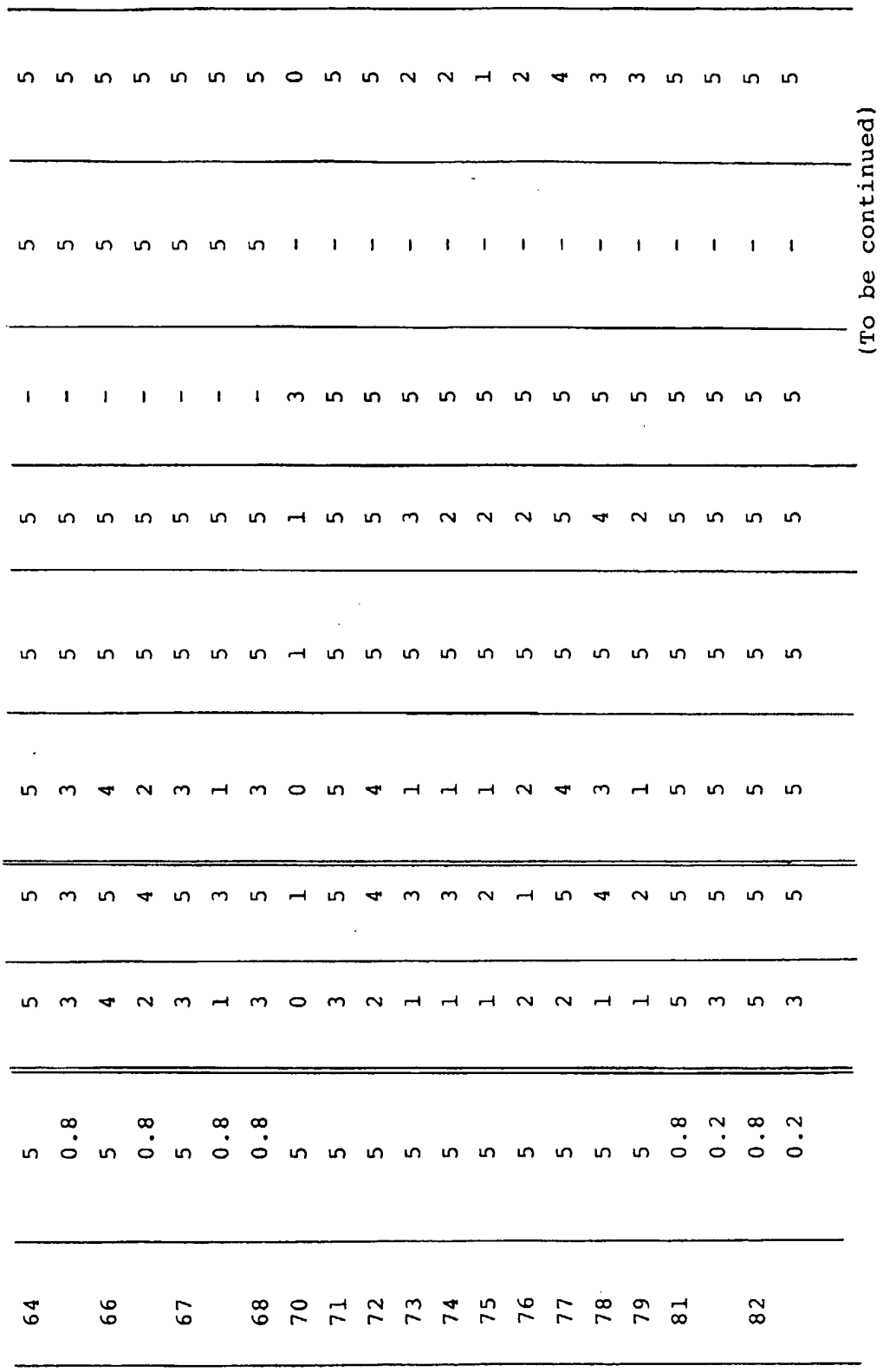

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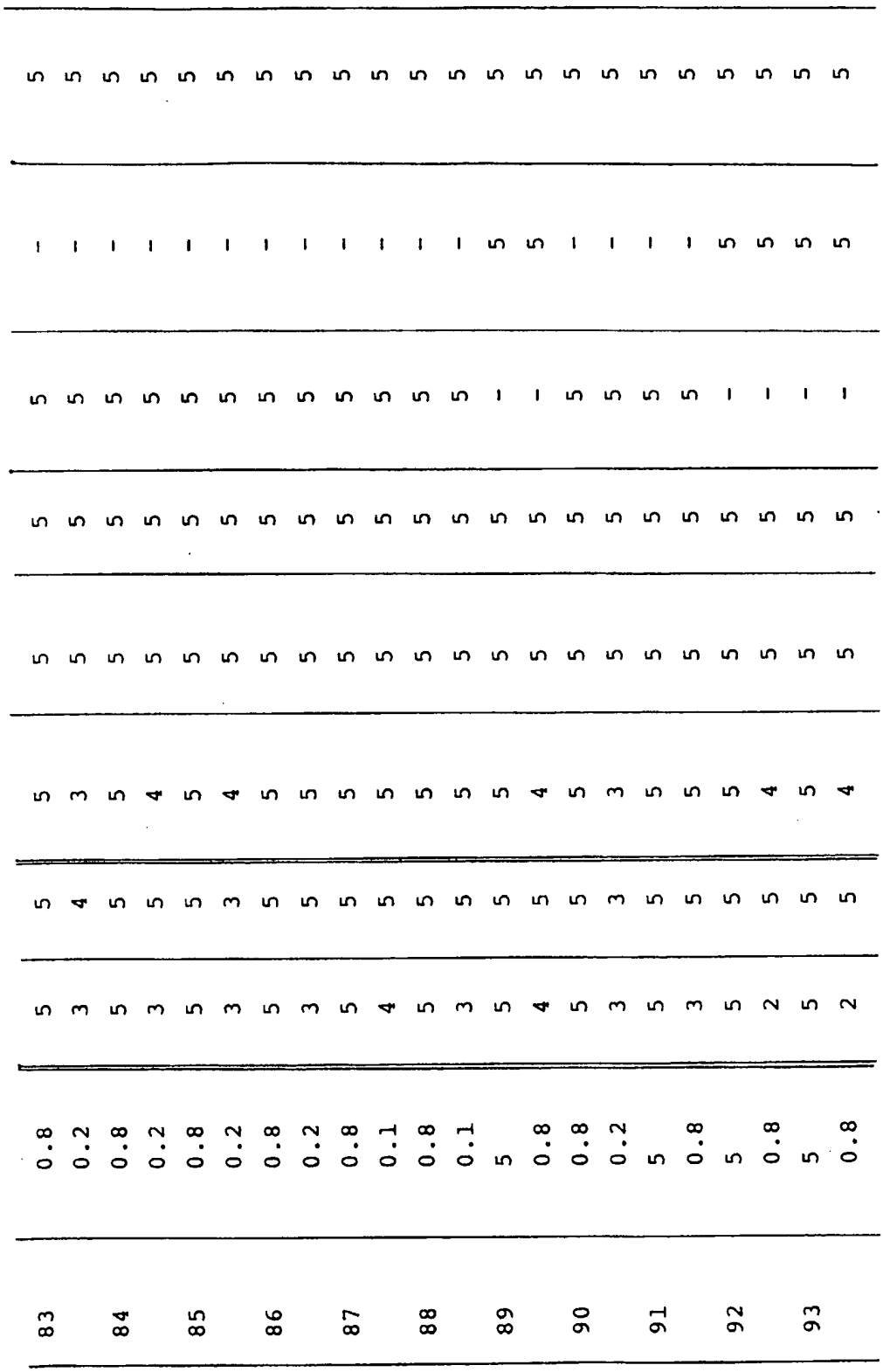

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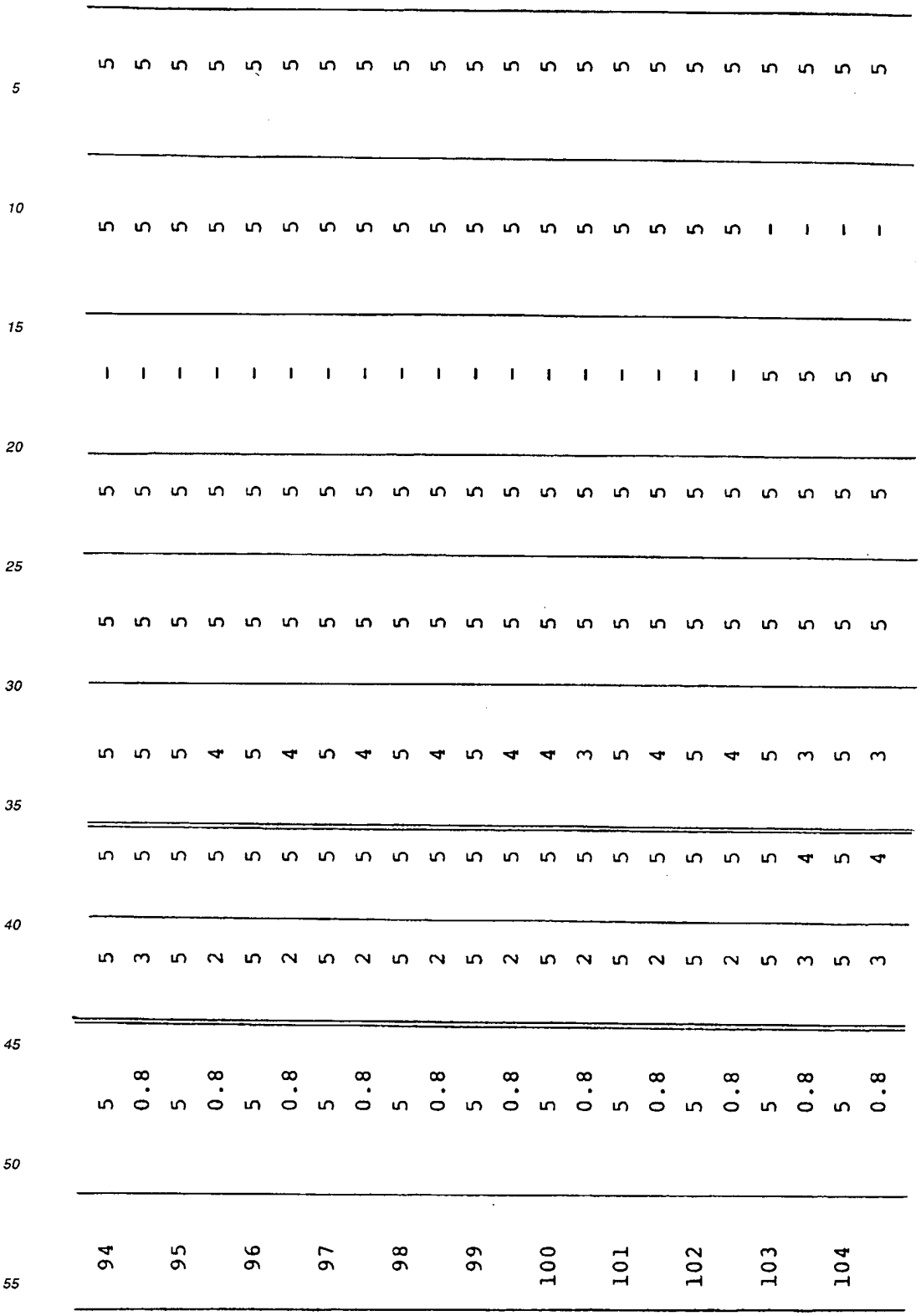

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107	-	-	5	5	5	5	5	5	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
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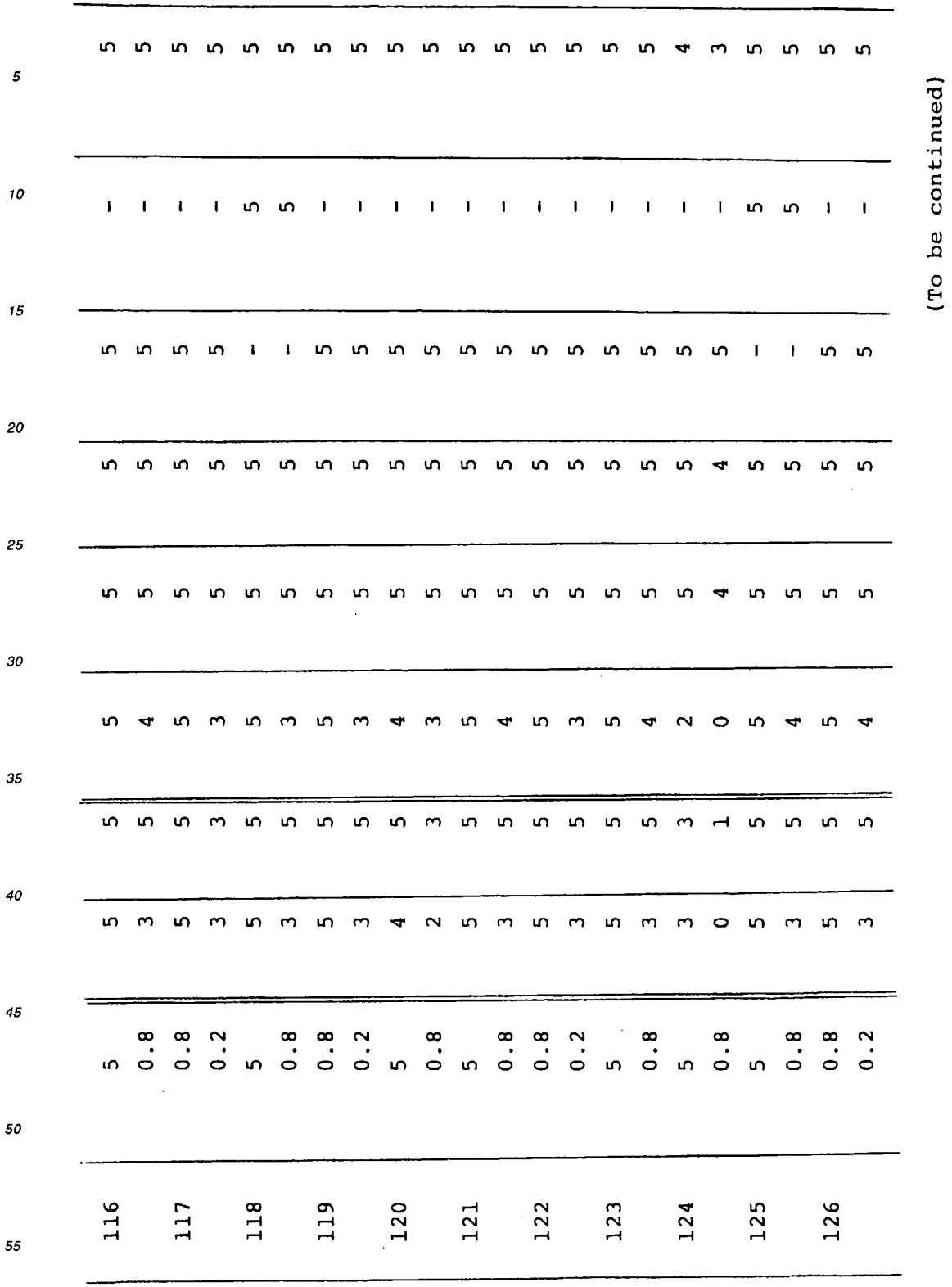

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123	5	0.2	5	3	5	3	5	3	4	2	5	3	5	3	5	3	3	0	5	3	5	5	5	3
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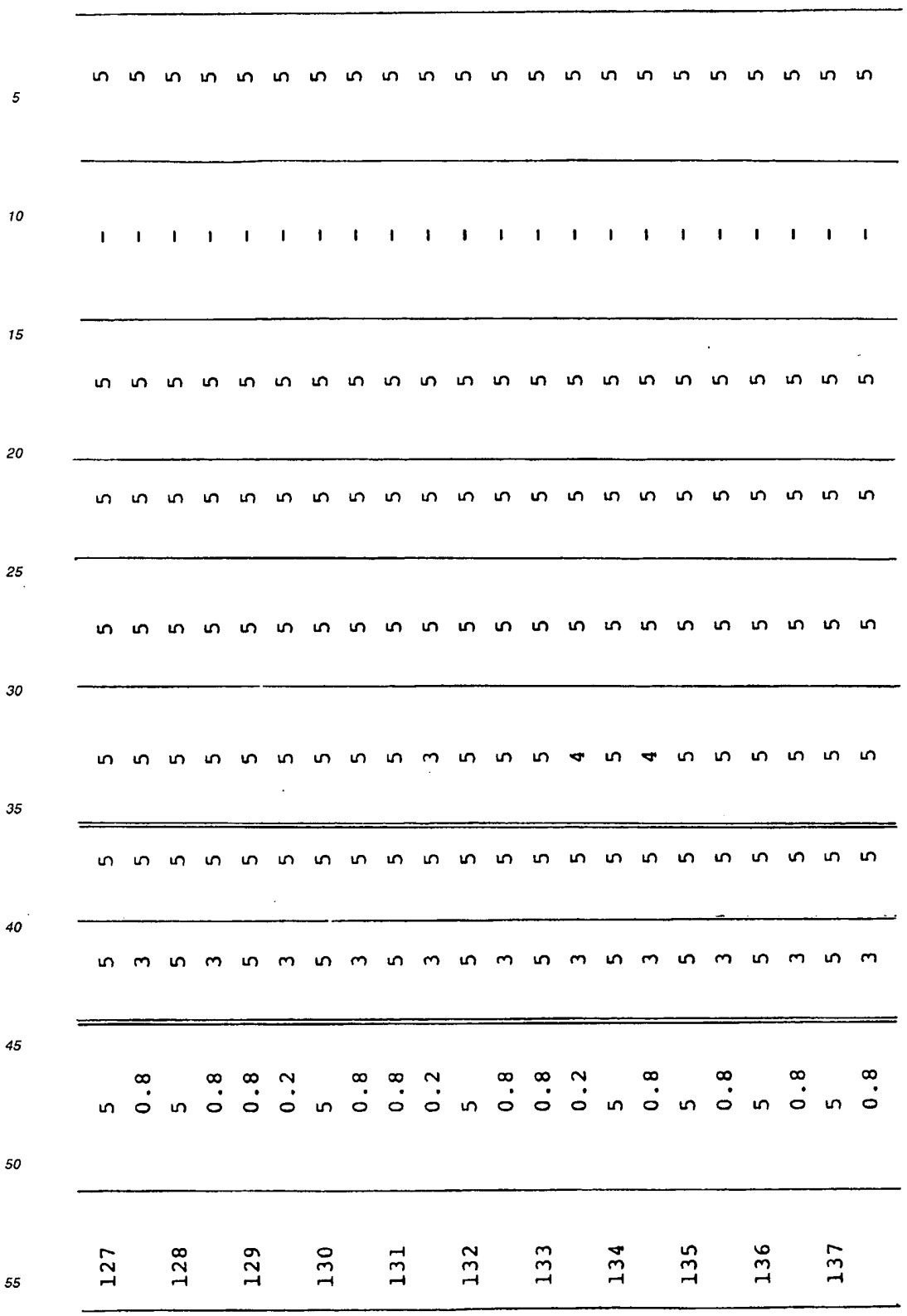

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149																								
150																								

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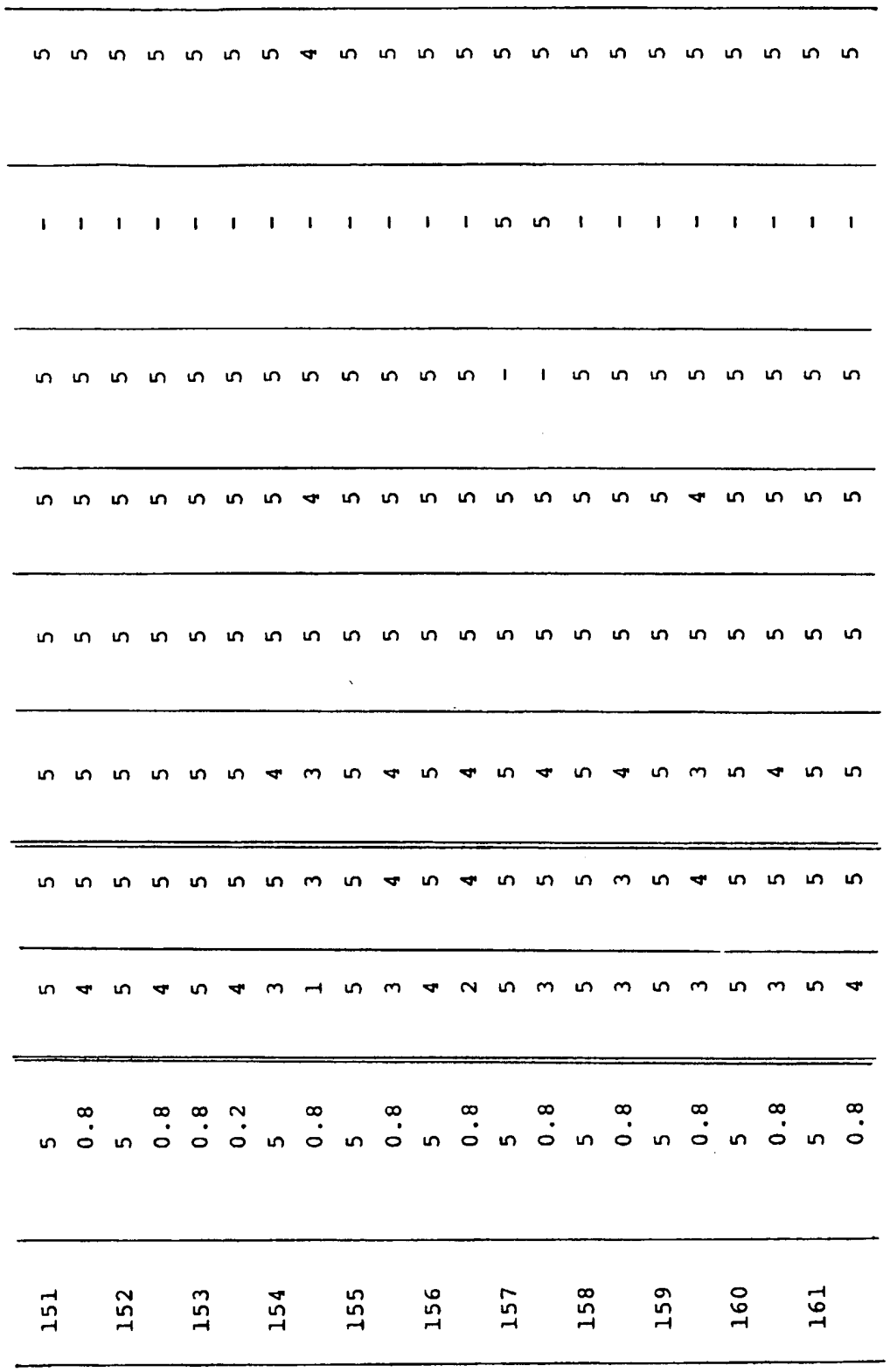

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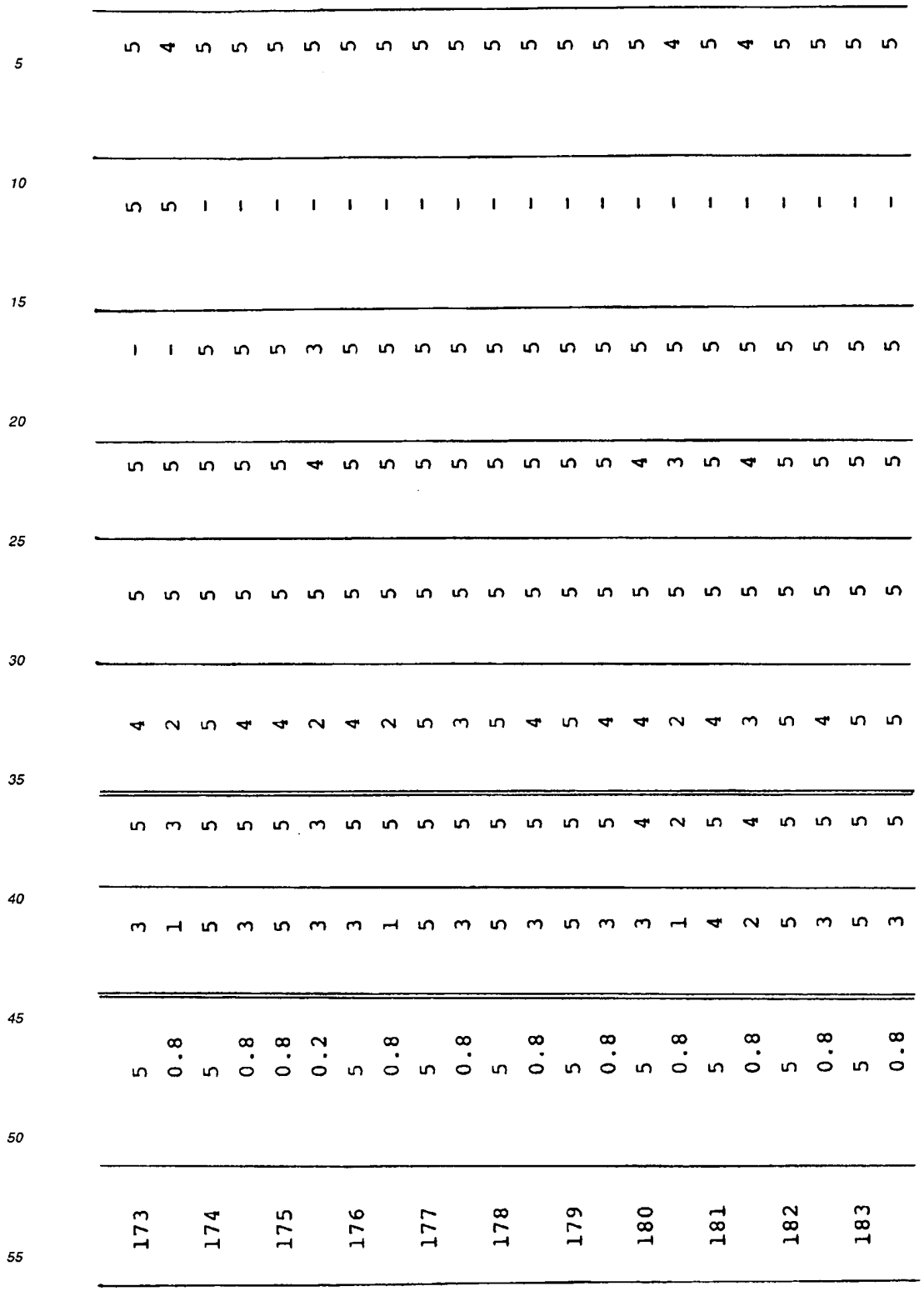

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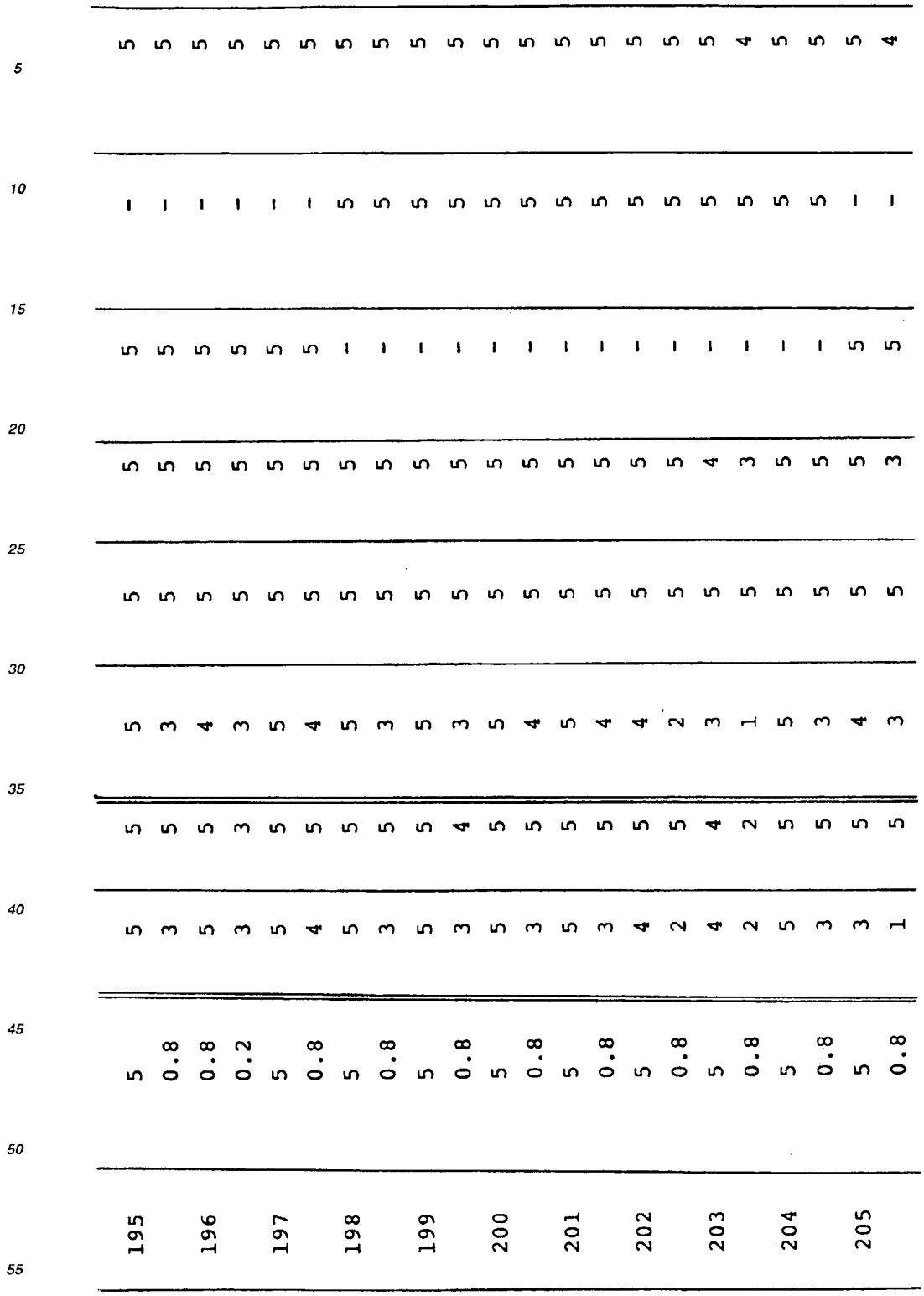

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204																							
205																							

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215	5	2	5	3	5	5	5	-	5
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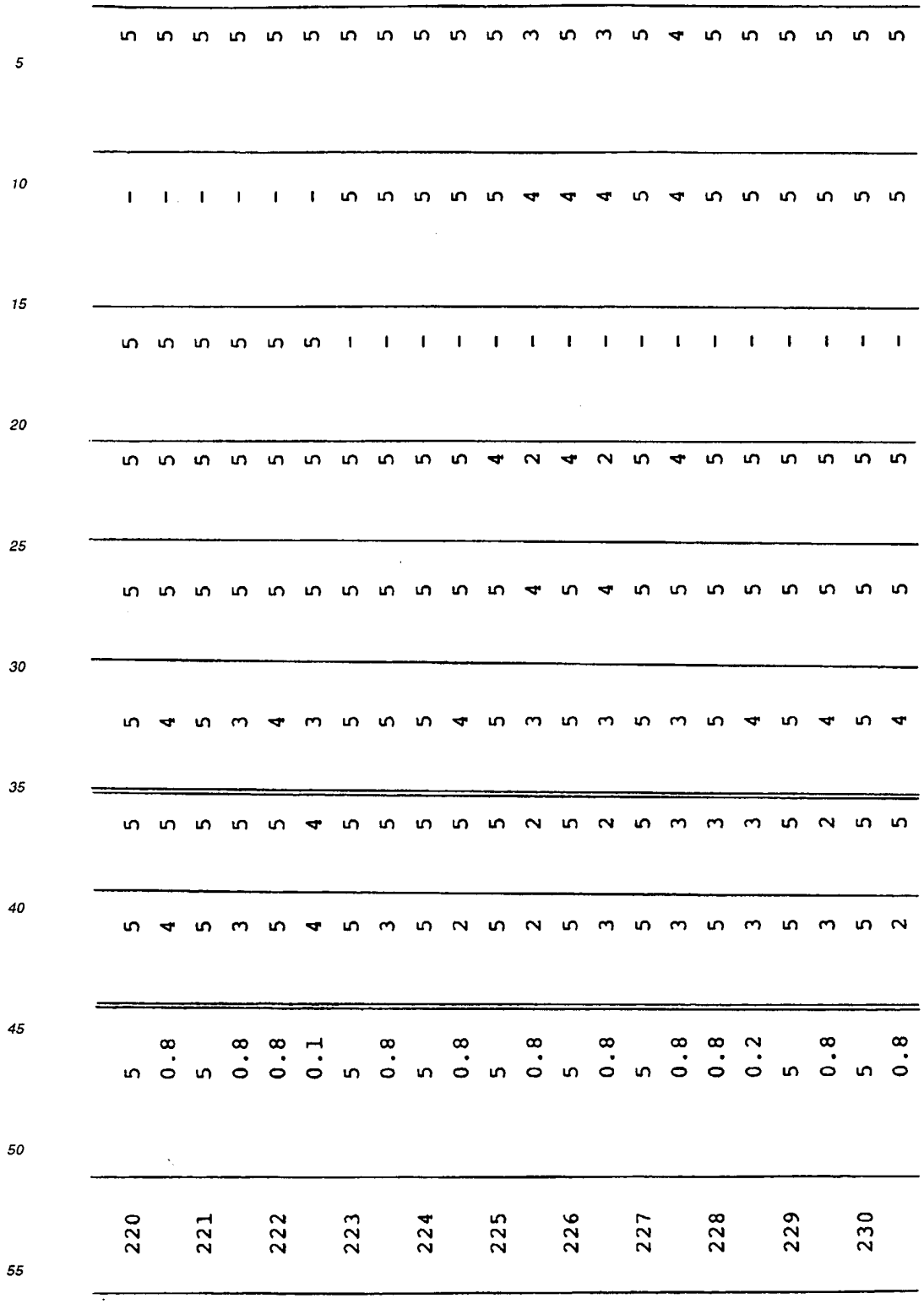

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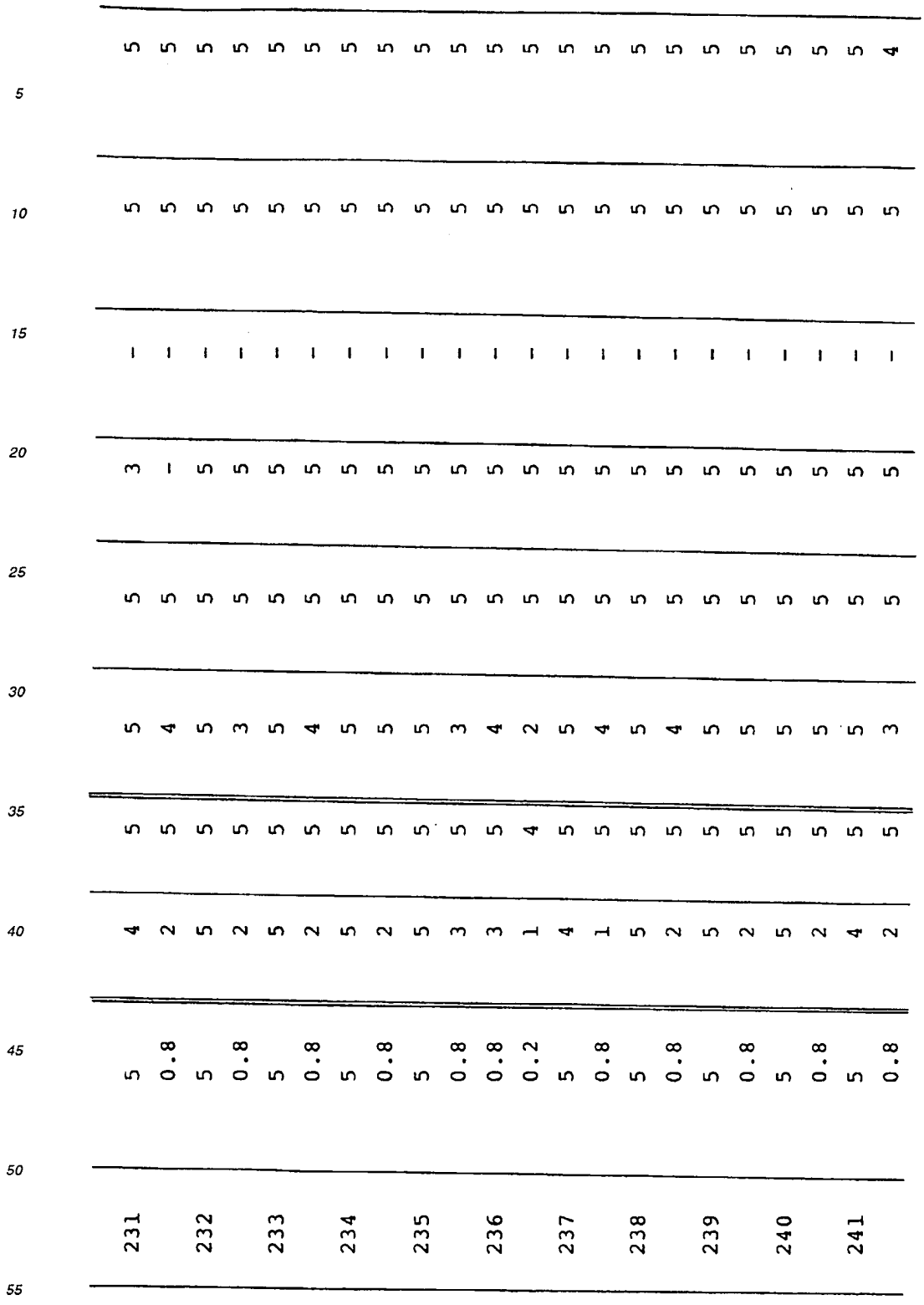

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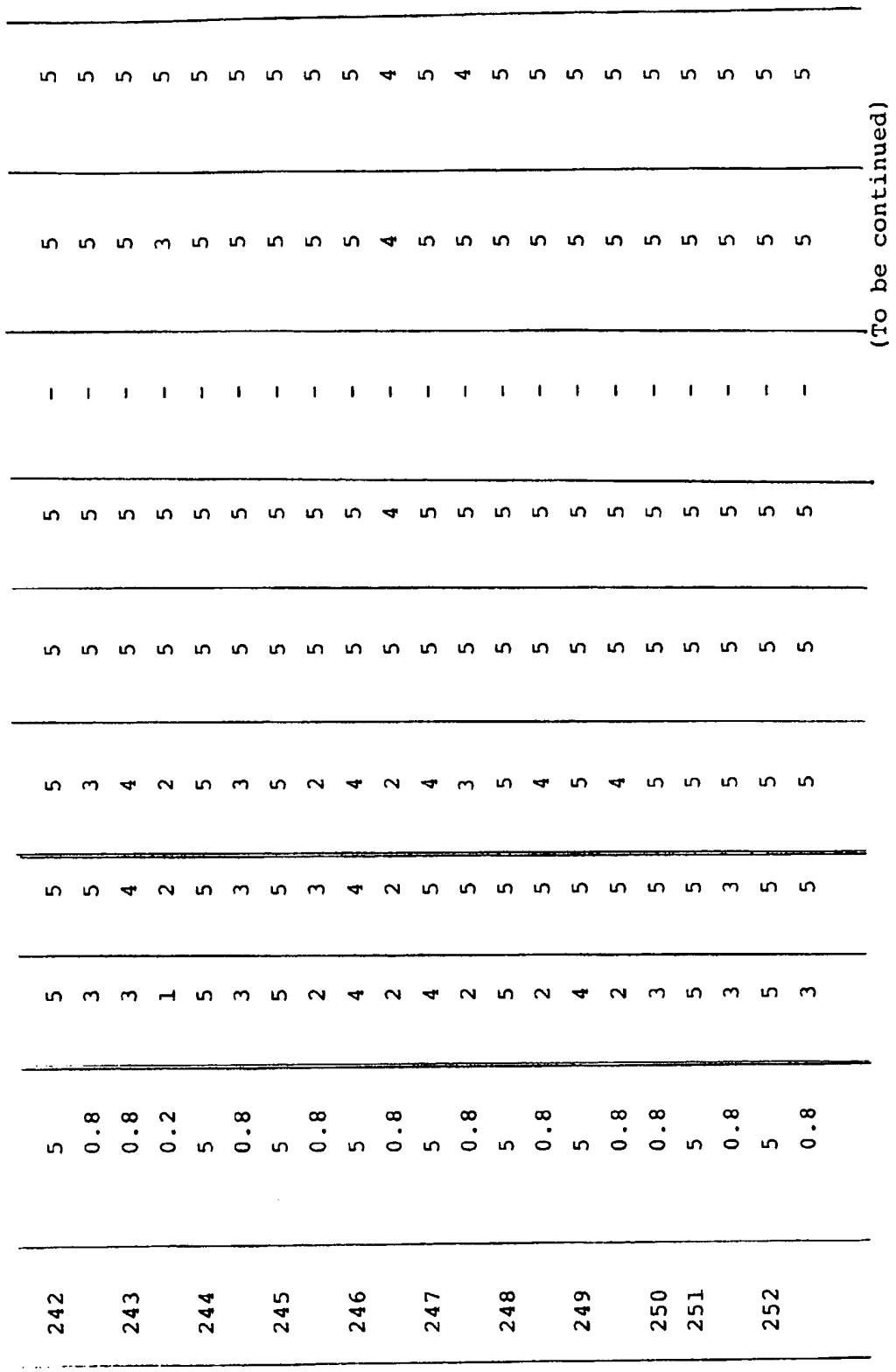

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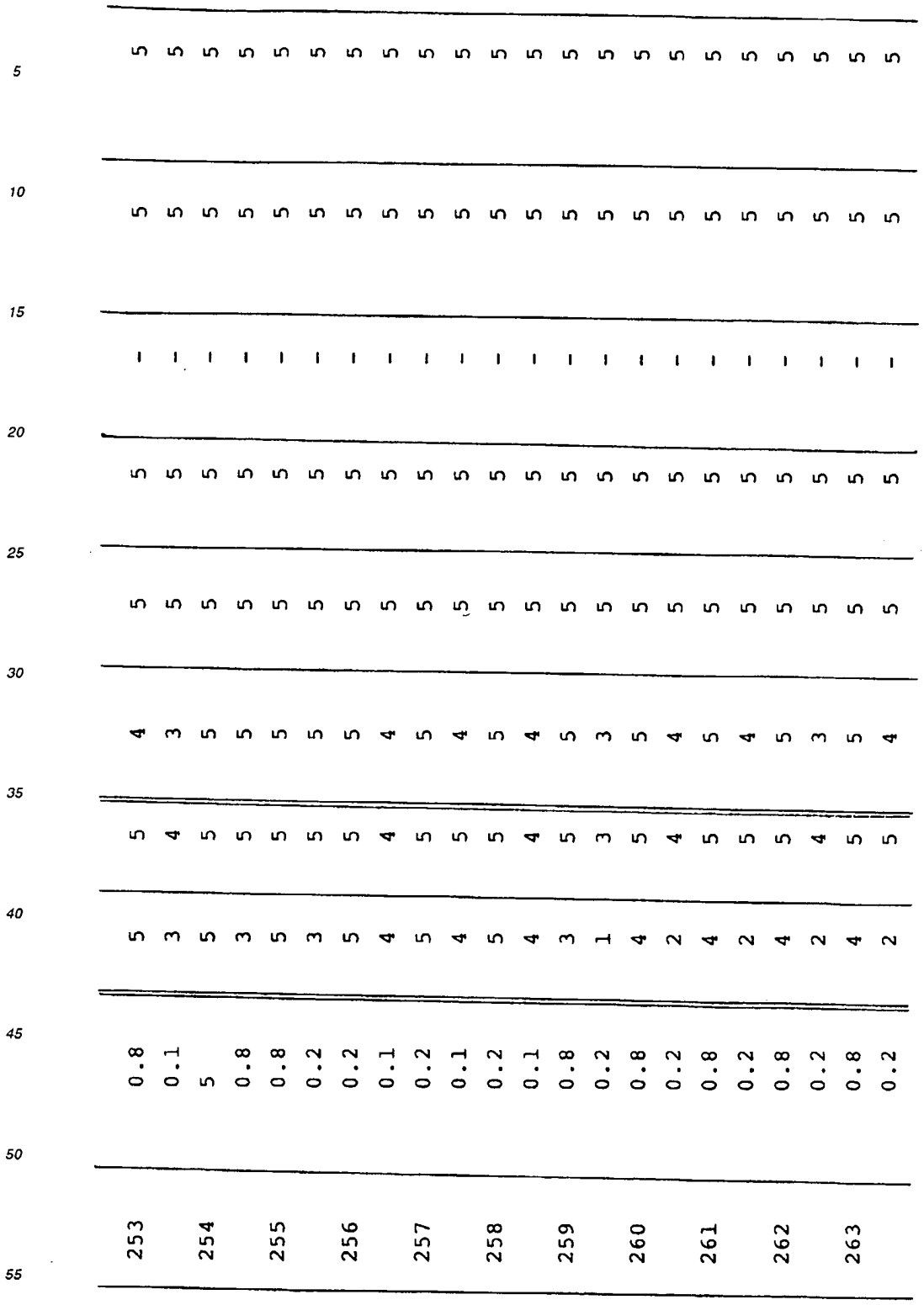

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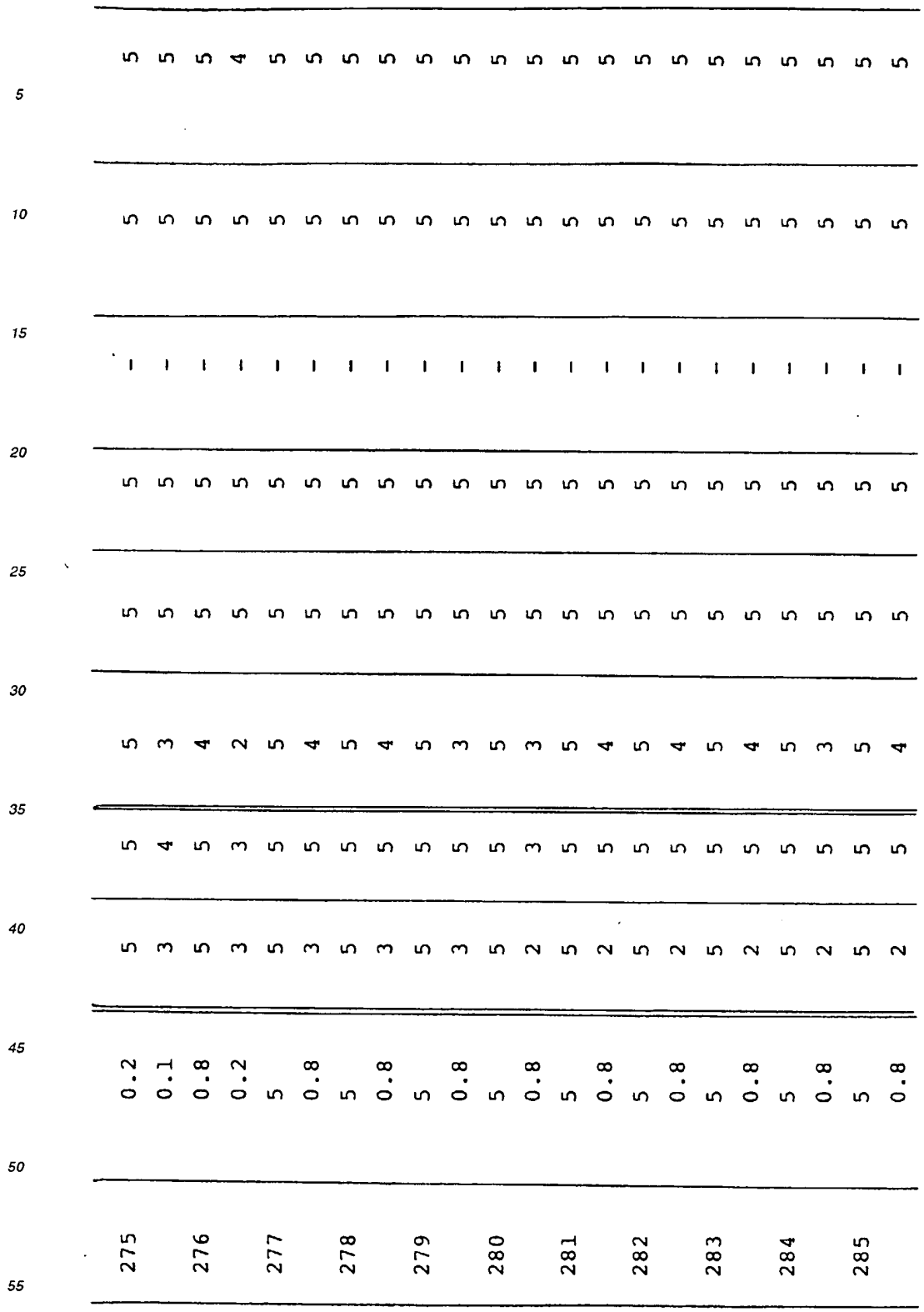

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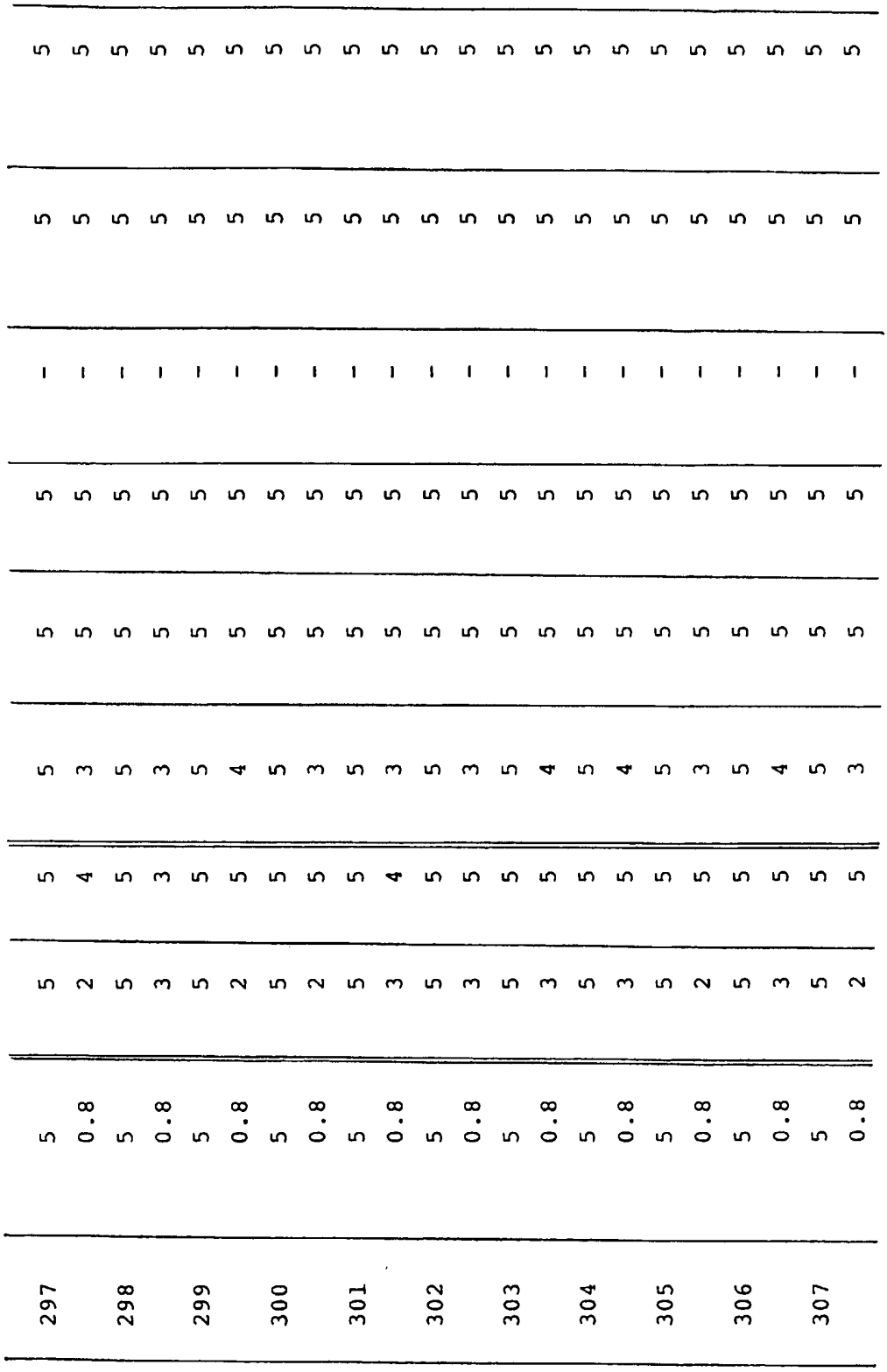

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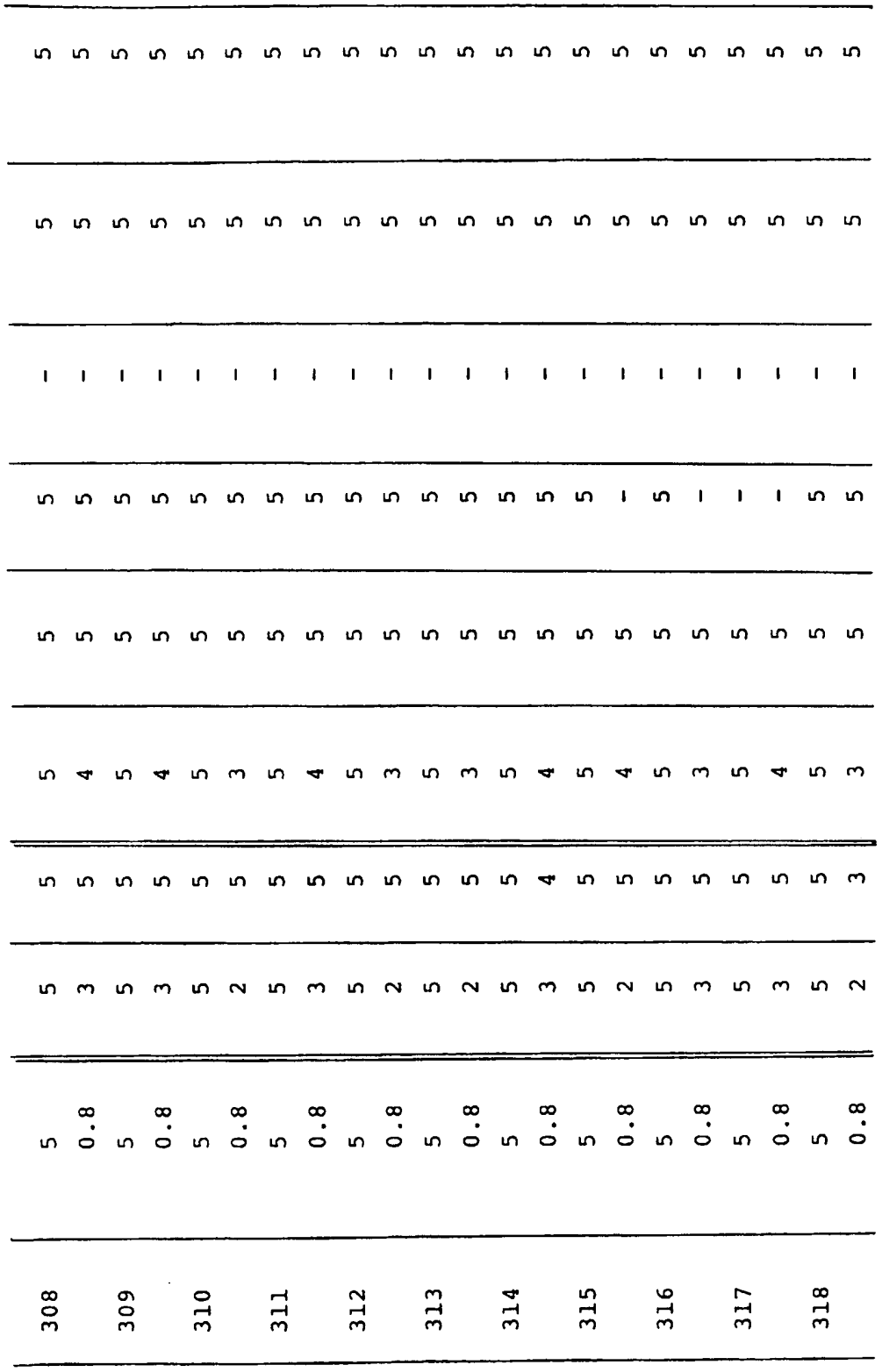

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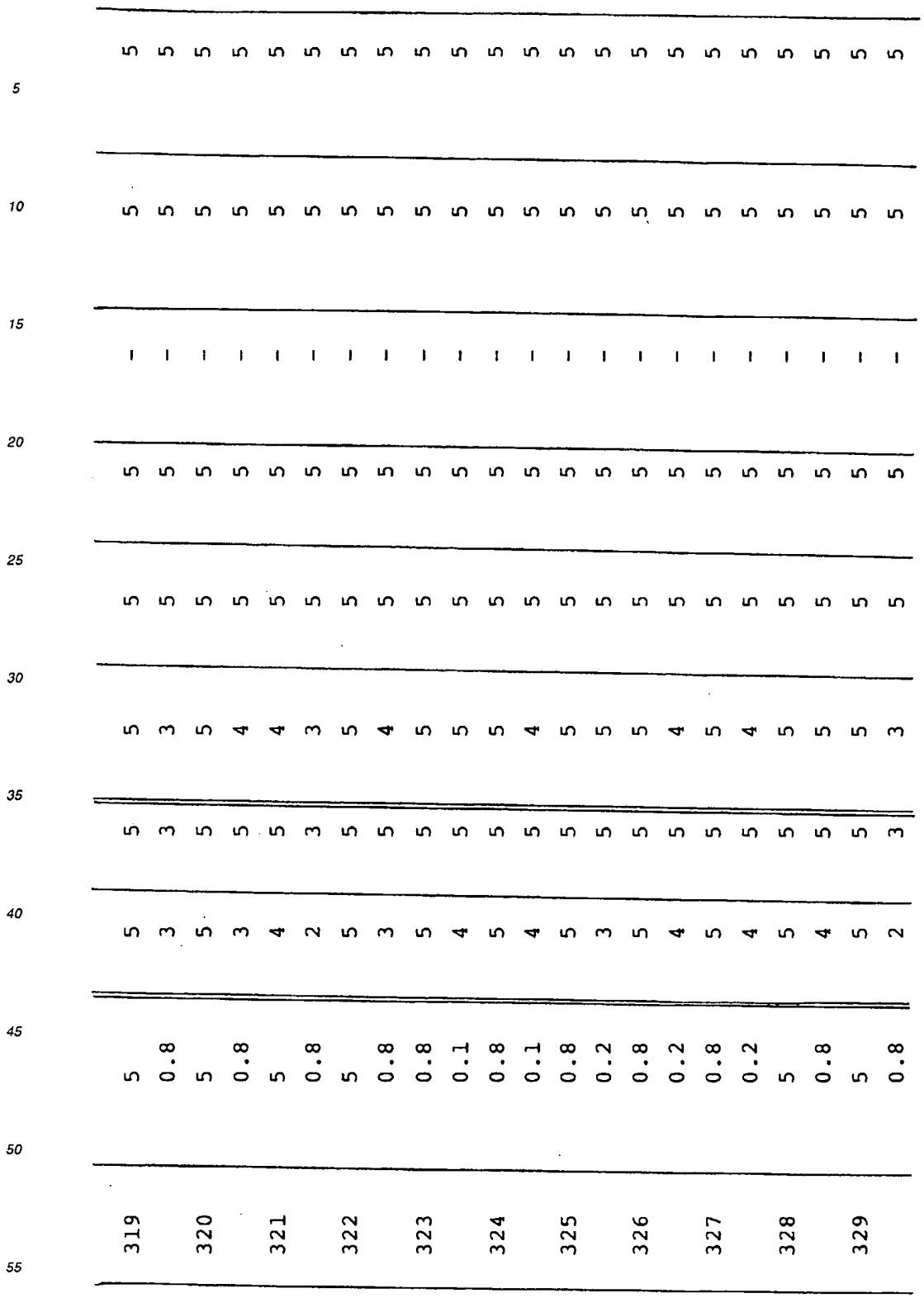

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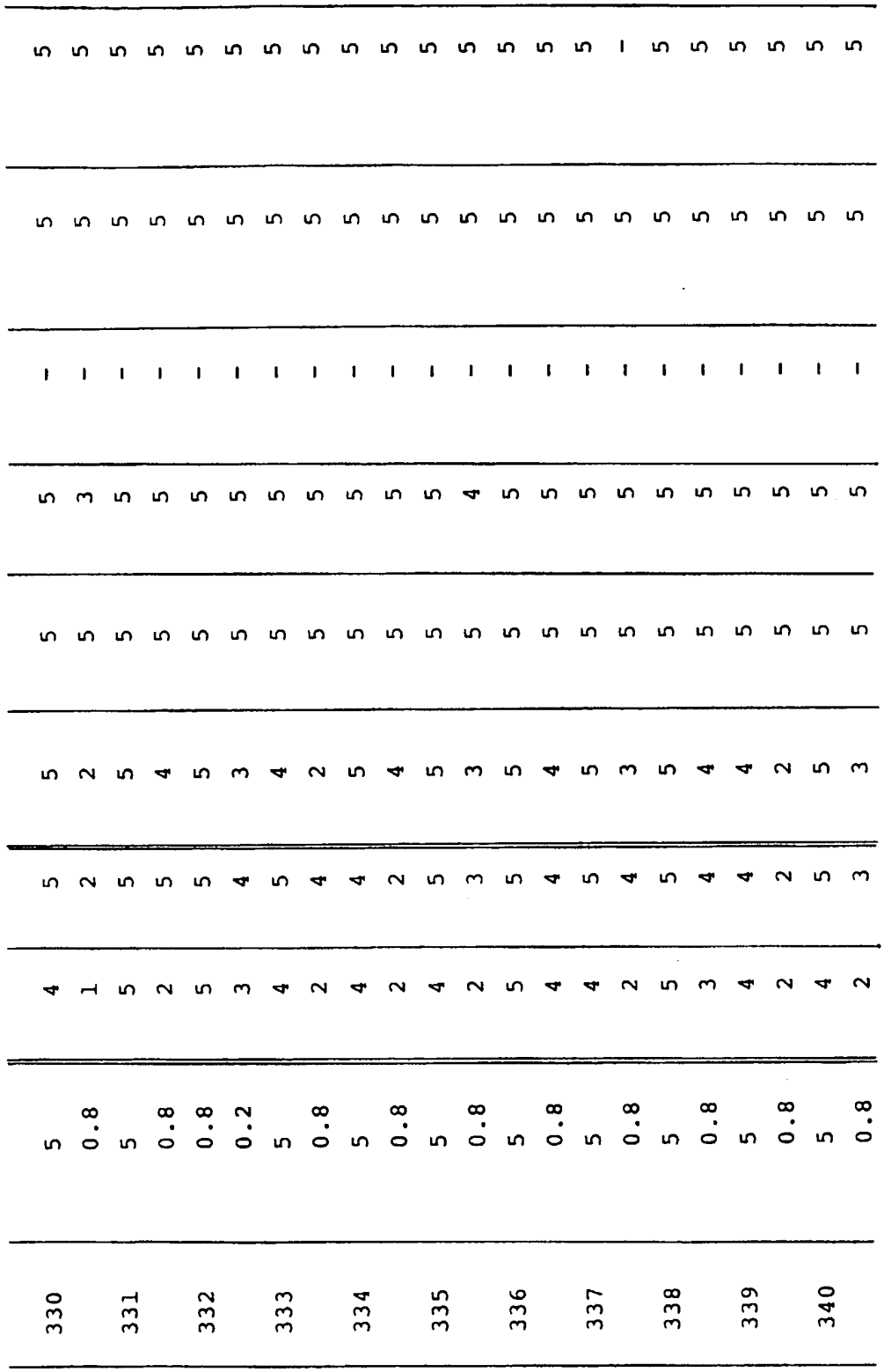

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334	0.8	4	4	5	2	4	5	5	5	5
335	0.2	3	5	5	3	4	4	4	4	4
336	0.8	4	5	5	4	5	5	5	5	5
337	0.8	2	4	5	4	5	5	5	5	5
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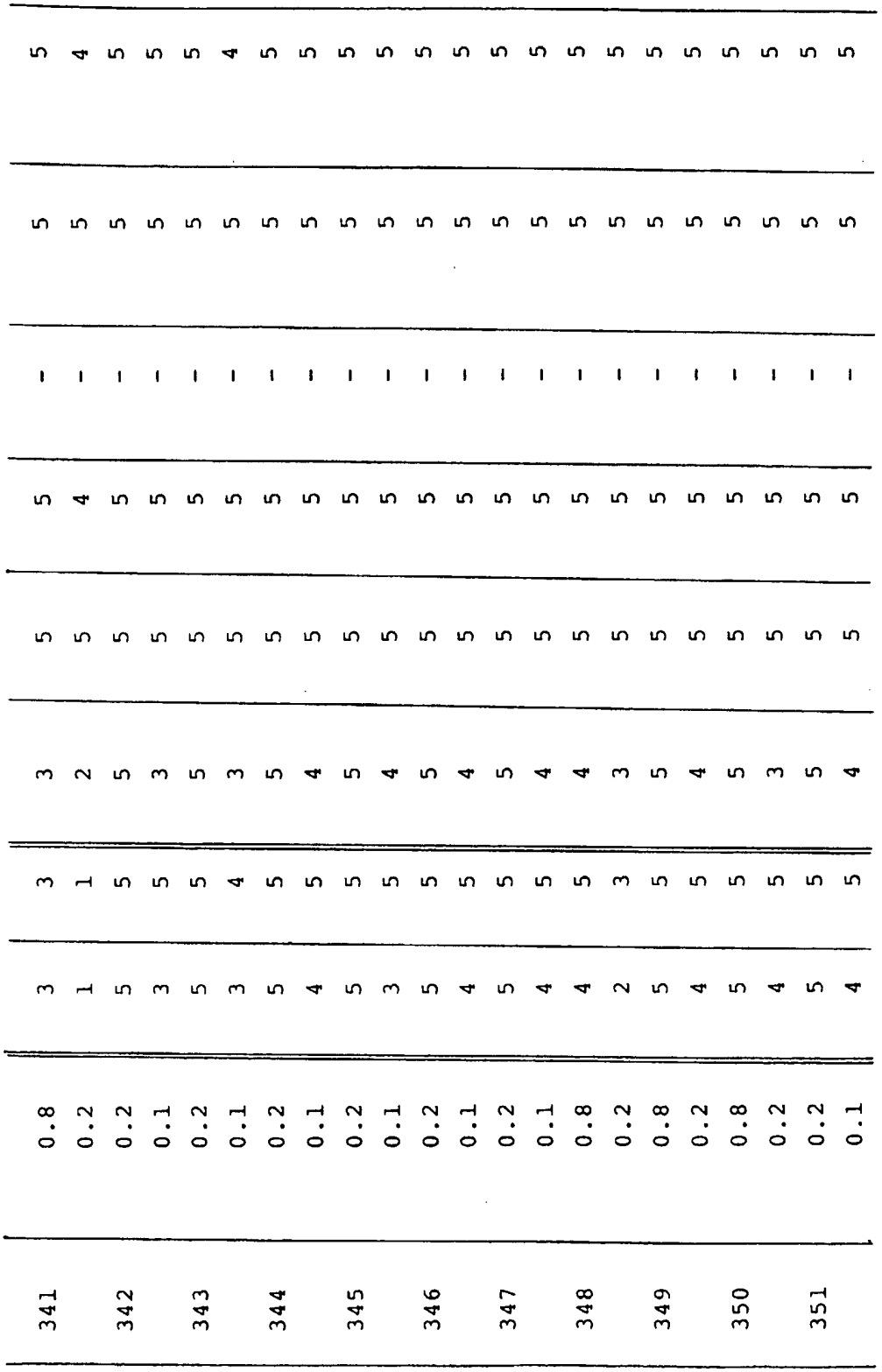

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(To be continued)

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(To be continued)

363	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
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369	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
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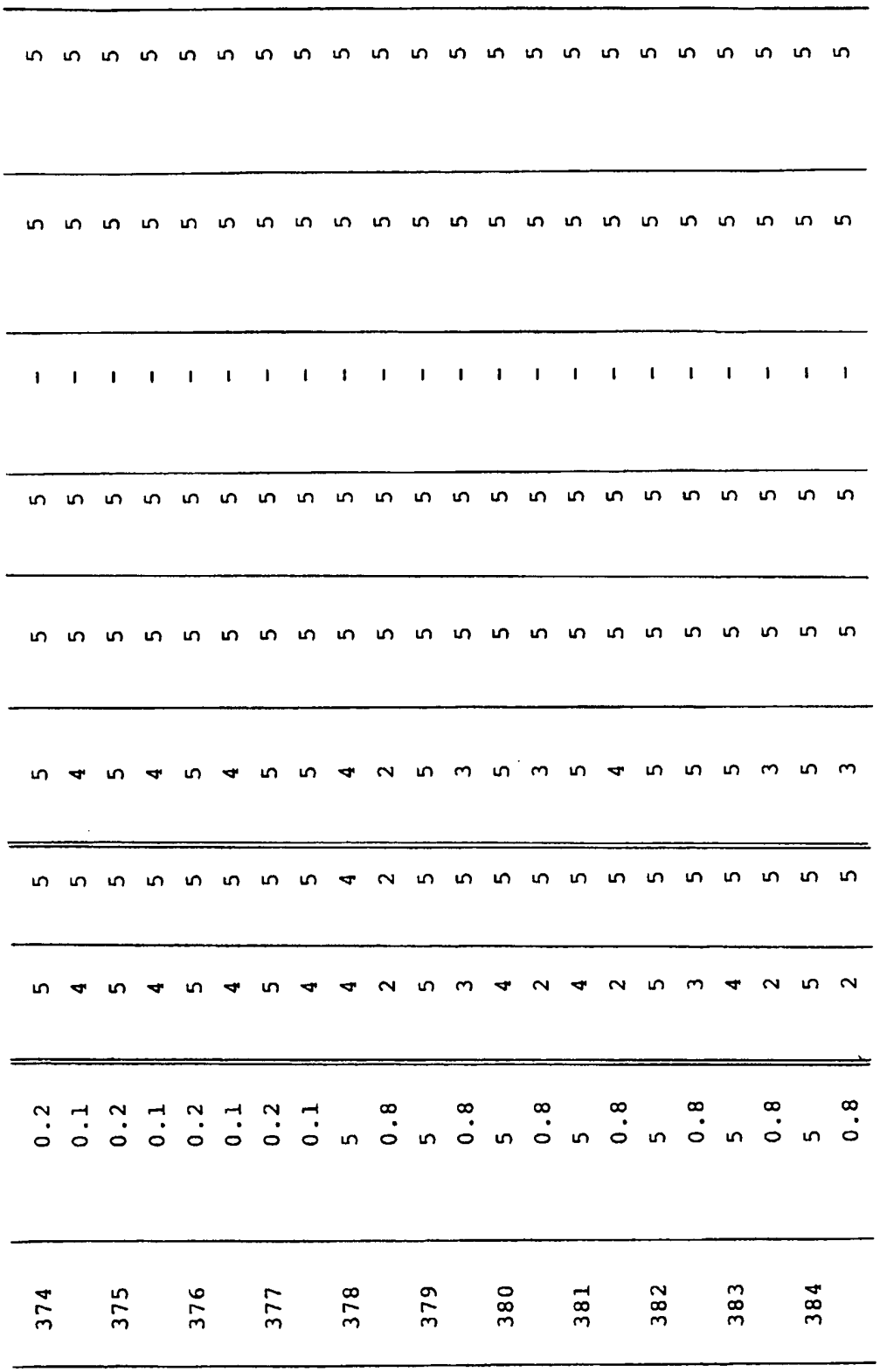

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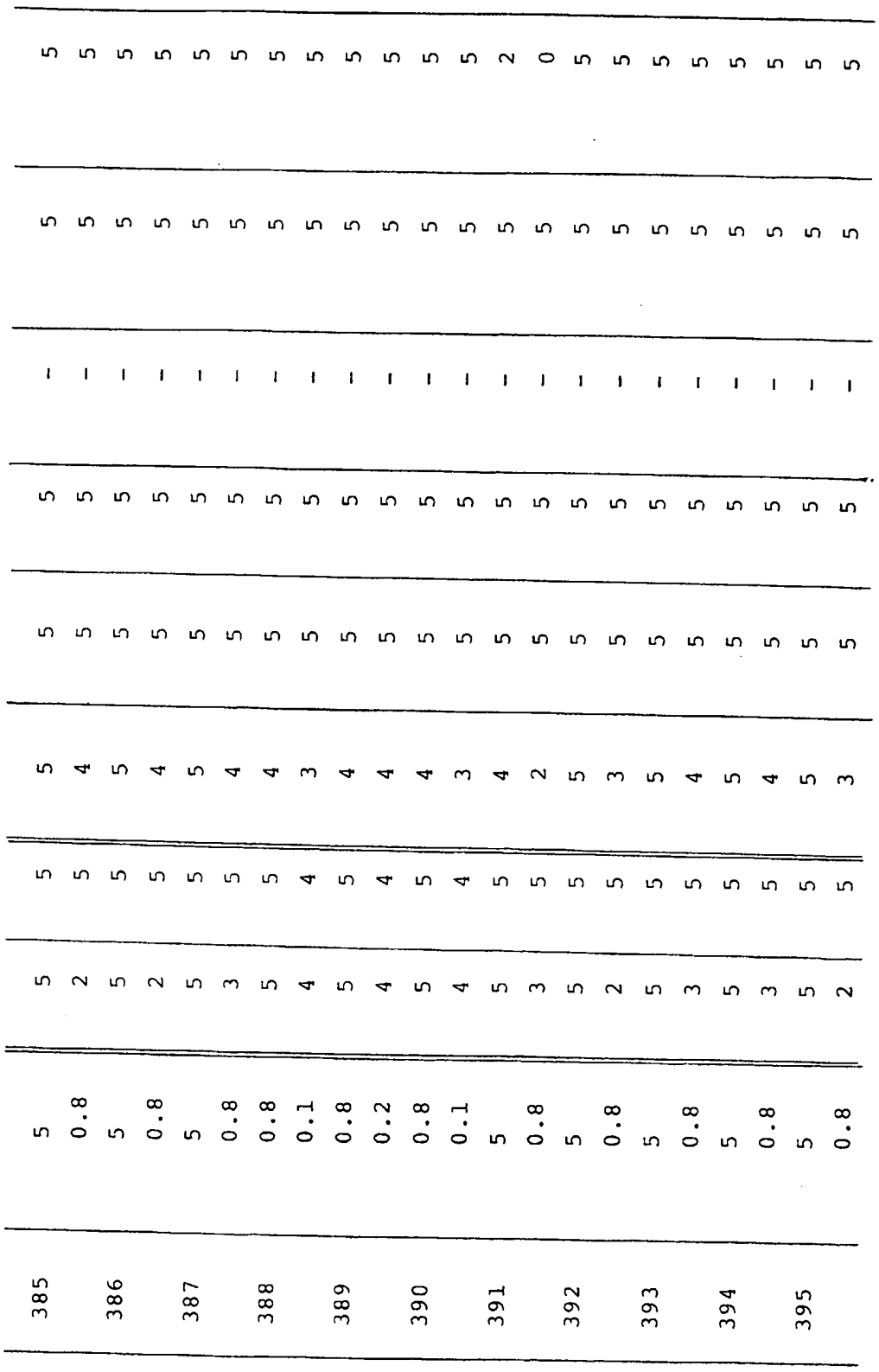
(To be continued)



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392	5	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
393	5	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
394	5	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
395	5	0.8	0.8	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

(To be continued)

396	5	4	5	4	5	5	-	5	5
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397	5	5	5	5	5	5	-	5	5
	0.8	3	4	2	5	5	-	5	5
398	5	4	5	4	5	5	-	5	5
	0.8	2	5	2	5	5	-	5	5
399	5	5	5	5	5	5	-	5	5
	0.8	2	5	3	5	5	-	5	5
400	0.8	5	5	4	5	5	-	5	5
	0.2	4	5	4	5	5	-	5	5
401	5	5	5	5	5	5	-	5	5
	0.8	2	5	4	5	5	-	5	5
402	5	5	5	5	5	5	-	5	5
	0.8	2	5	4	5	5	-	5	5
403	0.8	5	5	4	5	5	-	5	5
	0.2	4	5	3	5	5	-	5	5
404	5	5	5	4	5	5	-	5	5
	0.8	3	4	3	5	5	-	5	5
405	5	5	5	4	5	5	-	5	5
	0.8	3	4	3	5	5	-	5	5
406	5	5	5	4	5	5	-	5	5
	0.8	3	4	3	5	4	-	5	5

(To be continued)

A	5	0	2	0	3	1	2	3	-	3	
B	5	0	1	0	2	0	3	2	-	2	
C	5	0	1	0	1	0	1	1	-	1	
D	5	0	2	0	3	1	3	3	-	3	

As shown in table 8, the present invented 3-(substituted phenyl)pyrazole derivatives or salt thereof, represented

by the general formula (I), exhibit more excellent controlling effect on weeds than comparative compound A, B, C or D on post-emergence treatment in upland fields. Even when the invented derivatives exhibit both herbicidal activity on weeds and phytotoxicity on crops at same dosage, by the selection of proper lower dosage, sufficient herbicidal activity is retained and chemical injury on crops is diminished remarkably.

Formulation Example 1

A wettable powder composition was prepared by uniform mixing and grinding of the following ingredients:

Compound No. 1	50 parts
Clay-white carbon mixture (clay is the major component)	45 "
Polyoxyethylene nonyphenyl ether	5 "

Formulation Example 2

A granular composition was prepared by uniform mixing and grinding of the following ingredients, kneading the mixture with a suitable amount of water, and granulating the kneaded mxture.

Compound No. 7	5 parts
Bentonite-clay mixture	90 "
Calcium ligninsulfonate	5 "

Formulation Example 3

An emulsifiable concentrate was prepared by uniform mixing of the following ingredients:

Compound No. 12	50 parts
Xylene	40 "
Mixture of polyoxyethylene nonylphenyl ether and calcium alkylbenzenesulfonate	10 "

Formulation Example 4

A wettable powder composition was prepared by uniform mixing and grinding of the following ingredients:

Compound No. 33	50 parts
Clay-white carbon mixture (clay is the major component)	45 "
Polyoxyethylene nonylphenyl ether	5 "

Formulation Example 5

A granular composition was prepared by uniform mixing and grinding of the following ingredients, kneading the mixture with a suitable amount of water, and granulating the kneaded mixture.

Compound No. 122	5 parts
Bentonite-clay mixture	90 "
Calcium ligninsulfonate	5 "

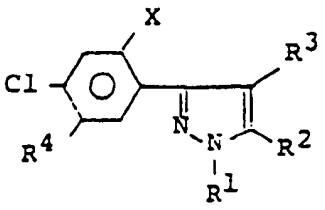
Formulation Example 6

An emulsifiable concentrate was prepared by uniform mixing the following ingregients:

Compound No. 381	50 parts
Xylene	40 "
Mixture of polyoxyethylene nonylphenyl ether and calcium alkylbenzenesulfonate	10 "

Claims

1. A 3-(substituted phenyl)pyrazole derivative or a salt thereof, the derivative being represented by the general formula

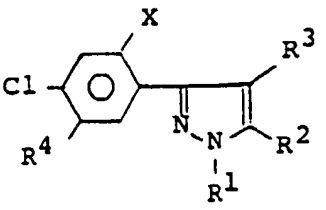


(I)

wherein

X denotes halogen,
R^1 denotes CH_3,
R^2 denotes hydroxy, mercapto, alkylthio, haloalkoxy or haloalkylthio,
R^3 denotes hydrogen or halogen, and
R^4 denotes (i) formyl, (ii) nitro, (iii) -CO-B-R^6 wherein B denotes -0-, -S-, or -NR^7, R^6 and R^7 are the same or different and each denote hydrogen or cycloalkyl, and when B is -O-, R^6 can be an alkali metal atom or -CO-B-R^6 denotes methoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, butoxycarbonyl, methoxycarbonylmethoxycarbonyl, ethoxycarbonylmethoxycarbonyl, propoxycarbonylmethoxycarbonyl, methoxycarbonylethoxycarbonyl, ethoxycarbonylethoxycarbonyl, or propoxycarbonylethoxycarbonyl,
(iv) -D-R^8 wherein D denotes -O-,-$S(O)_n$, n being an integer of 0 to 2, or -NR^9-, R^8 and R^9, which are the same or different, each denotes hydrogen; alkyl; haloalkyl; aminosulfonyl; phenylalkyl or phenoxy-alkyl optionally having on the phenyl ring one or more halogen substituents which are the same or different; or -D-R^8 denotes an alkenyloxy group containing propenyl, butenyl, or pentenyl; an alkynyloxy group containing propynyl,butynyl or pentynyl; an alkenylamino group containing propenyl, butenyl or pentenyl; an alkoxy-carbonylalkoxy or an alkylthiocarbonylalkoxy group containing each methyl, ethyl, propyl or butyl; or (v) -$(CHR^{10})_m$-CO-E-R^{12} wherein E denotes -0-, -S-, or -NR^{11}; R^{10} denotes hydrogen and R^{11} and R^{12}, which are the same or different, each denotes hydrogen; alkyl; haloalkyl; phenyl optionally having on the phenyl ring one or more halogen substituents; phenylalkyl optionally having on the phenyl ring one or more halogen substituents; or R^{11}, jointly with R^{12}, forms a piperidino or morpholinyl group, and when E is -O-, R^{12} can be an alkali metal atom; and m denotes an integer of 0 to 3.

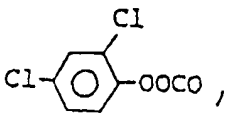
2. A 3-(substituted phenyl)-pyrazole derivative or a salt thereof, the derivative being represented by the general formula



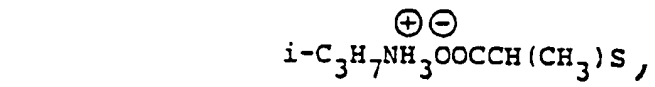
(I)

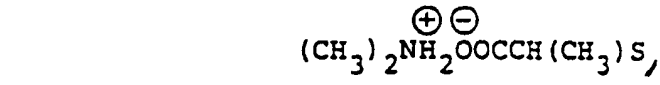
wherein

a) X is Cl, R^1 is CH_3, R^2 is CH_3S, R^3 is Cl and R^4 is selected from

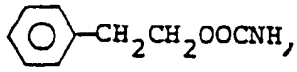


$HOOCCH(CH_3)O$, $CH_3OOCCH(CH_3)O$, $C_2H_5OOCCH(CH_3)O$, $i\text{-}C_3H_7OOCCH(CH_3)O$, $CH_2{=}CHCH_2OOCCH(CH_3)O$, $CH{\equiv}CCH_2OOCCH(CH_3)O$, $CH_2{=}CHCH_2S$, $CH_2{=}C(Cl)CH_2S$, $CH{\equiv}CCH_2S$, $HO_2CCH(CH_3)S$,





$Cl(CH_2)_3OOCCH(CH_3)S$, $CH_2{=}CHCH_2OOCCH(CH_3)S$, $CH{\equiv}CCH_2OOCCH(CH_3)S$, $C_2H_5CH(CH_3)CH_2OOCCH(CH_3)S$, $(C_2H_5)_2NCOCH(CH_3)S$, CH_3NHSO_2, $(CH_3)_2NSO_2$, $n\text{-}C_3H_7NHSO_2$, $CH_3SO_2NHSO_2$, $Cl^{\ominus}H_3N^{\oplus}$, $CH{\equiv}CCH_2NH$, CH_3OOCNH, CH_3SO_2NH, CH_3NHSO_2NH, $CH_3NHSO_2N(CH_3)$, $(CH_3)_2NSO_2N(CH_3)$, $i\text{-}C_3H_7NHSO_2NH$, $CH{\equiv}CCH_2N(CH_3)SO_2N(CH{\equiv}CCH_2)$, $CH_3OCH_2CH_2OOCNH$, $BrCH_2CH_2OOCNH$, $NCCH_2CH_2OOCNH$, $CH_2{=}C(CH_3)CH_2OOCNH$,



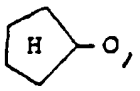
$CH_2{=}CHCH_2OOCCH_2NH$, $CH{\equiv}CCH_2OOCCH_2NH$, $C_2H_5OOCCH(CH_3)NH$, $HOOCCH_2NH$, CH_3OOCCH_2NH, $HOOCCH(CH_3)NH$ and $CH_3OOCCH(CH_3)NH$;

b) X is Cl, R^1 is CH_3, R^2 is CH_3SO, R^3 is Cl and R^4 is selected from NO_2, CH_3SO_2NH, CH_3NHSO_2NH, $i\text{-}C_3H_7NHSO_2NH$ and $CH_3NHSO_2(CH{\equiv}CCH_2)$;

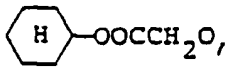
c) X is Cl, R^1 is CHF_2, R^2 is CH_3S, R^3 is Cl and R^4 is selected from NH_2, CH_3NHSO_2NH, $CH_3NHSO_2N(CH{\equiv}CCH_2)$ and $CH_3N(CH{\equiv}CCH_2)SO_2N(CH{\equiv}CCH_2)$;

d) X is Cl, R^1 is CHF_2, R^2 is CH_3SO, R^3 is Cl and R^4 is selected from NH_2 and NO_2;

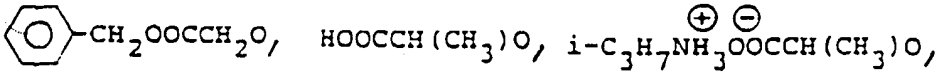
e) X is Cl, R^1 is CH_3, R^2 is CHF_2O, R^3 is Cl and R^4 is selected from



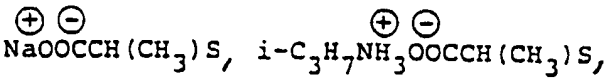
$C_2H_5OCH_2CH_2O$, $NCCH_2O$, CH_3OOCO, $n\text{-}C_6H_{13}OOCCH_2O$, $CH_2{=}CHCH_2OOCCH_2O$, $CH{\equiv}CCH_2OOCCH_2O$,



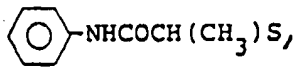
$C_2H_5O(CH_2)_2OOCCH_2O$, $C_2H_5O(CH_2)_2O(CH_2)_2OOCCH_2O$,



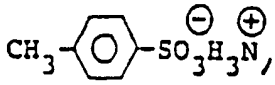
CH_2=$CHCH_2OOCCH(CH_3)O$, $CH{\equiv}CCH_2OOCCH(CH_3)O$, $CH_3NHCOCH(CH_3)O$, $(CH_3)_2NCOCH(CH_3)O$, CH_2=$CHCH_2S$, CH_2=$CHCH_2SO$, $CH{\equiv}CCH_2S$, $(C_2H_5O)_2P(O)CH_2S$, CH_3OOCS, CH_3OOCCH_2S, $C_2H_5OOCCH_2S$, CH_2=$CHCH_2OOCCH_2S$, $HOOCCH(CH_3)S$,



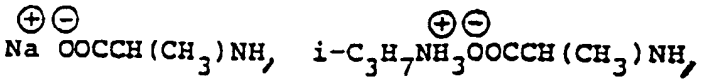
$CH_3OOCCH(CH_3)S$, $C_2H_5OOCCH(CH_3)S$, $C_2H_5OOCCH(CH_3)S(O)$, $C_2H_5OOCCH(CH_3)SO_2$, i-$C_3H_7OOCCH(CH_3)S$, n-$C_4H_9OOCCH(CH_3)S$, $(CH_3)_2CH(CH_2)_2OOCCH(CH_3)S$, CH_2=$CHCH_2OOCCH(CH_3)S$, $CH{\equiv}CCH_2OOCCH(CH_3)S$, $CH_3O(CH_2)_2O(CH_2)_2OOCCH(CH_3)S$, $(C_2H_5O)_2P(O)CH_2OOCCH(CH_3)S$, $CH_3NHCOCH(CH_3)S$, $(CH_3)_2NCOCH(CH_3)S$, $(C_2H_5)_2NCOCH(CH_3)S$, i-$C_3H_7NHCOCH(CH_3)S$,



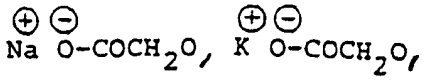
CH_3SO_2NH, CH_3NHSO_2NH, CH_2=$CHCH_2NH$, $CH{\equiv}CCH_2NH$, CH_3OOCNH, $Cl^{\ominus}H_3N^{\oplus}$,

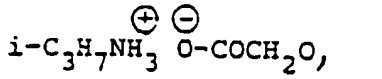


CH_3OOCCH_2NH, $HOOCCH(CH_3)NH$,

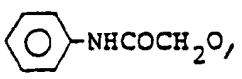


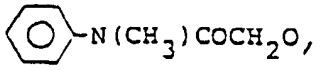
$C_2H_5OOCCH(CH_3)N$, $CH_3NHOCCH(CH_3)NH$, $(CH_3)_2NOCCH(CH_3)NH$, CH_3NHSO_2, $(CH_3)_2NSO_2$, $(C_2H_5)_2NCOS$, $(CH_3)_3SiCH_2S$, $(CH_3)_3SiCH_2O$, $HO\text{-}COCH_2O$,



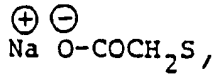


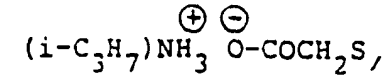
CH_2=$CHC(CH_3)_2O\text{-}COCH_2O$, $CH_3C{\equiv}CCH_2O\text{-}COCH_2O$, $CH_3NHCOCH_2O$, $(CH_3)_2NCOCH_2O$, n-$C_4H_9NHCOCH_2O$, H_2C=$CHCH_2NHCOCH_2O$, $(H_2C$=$CHCH_2)_2NCOCH_2O$,



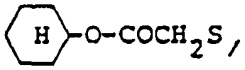


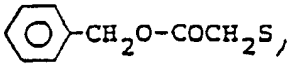
$CH_3SO_2NHCOCH_2O$, $CH_3SO_2NHCOCH(CH_3)O$, $(CH_3)_3SiCH_2O$-$COCH_2O$, $CH_3OC_2H_4OC_2H_4O$-$COCH(CH_3)O$, $CH_2=C(CH_3)CH_2S$, $CH_2=C(Cl)CH_2S$, $ClCH=CHCH_2S$, $ClCH=C(Cl)CH_2S$, $NCCH_2S$,



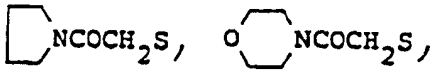


HO-$COCH_2S$, i-C_3H_7O-$COCH_2S$, n-C_4H_9O-$COCH_2S$, $C_{12}H_{25}O$-$COCH_2S$, CH_3S-$COCH_2S$, C_2H_5S-$COCH_2S$, i-C_3H_7S-$COCH_2S$, $CH{\equiv}CCH_2O$-$COCH_2S$, $C_2H_5OC_2H_4OC_2H_4O$-$COCH_2S$,

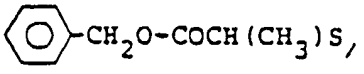




$(CH_3)_3SiCH_2O$-$COCH_2S$, $CH_3NHCOCH_2S$, $(CH_3)_2NCOCH_2S$, $H_2C=CHCH_2NHCOCH_2S$, $(H_2C=CHCH_2)_2NCOCH_2S$, $HC{\equiv}CCH_2NHCOCH_2S$,

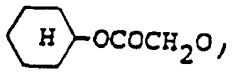


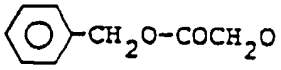
i-C_4H_9O-$COCH(CH_3)S$, t-$C_4H_9CH_2O$-$COCH(CH_3)S$, n-$C_6H_{13}O$-$COCH(CH_3)S$,



C_2H_5S-$COCH(CH_3)S$, i-C_3H_7S-$COCH(CH_3)S$, $NCCH(CH_3)O$-$COCH(CH_3)S$, $CH_3SC_2H_4O$-$COCH(CH_3)S$,

$(CH_3)_3SiCH_2O$-$COCH(CH_3)S$, CH_2=$CHCH_2NHCOCH(CH_3)S$, $(H_2C$=$CHCH_2)_2NCOCH(CH_3)S$, HC≡$CCH_2NHCOCH(CH_3)S$, CH_3CO-O-$(CH_2)_2S$, C_2H_5O-$COC(CH_3)_2S$, CH_3O-$COCH(i$-$C_3H_7)S$, C_2H_5O-$COCH(CH_3)N(CH_3)$, CH_2=$CHCH_2O$-CO, CH_3O-$COCH(CH_3)O$-CO, C_2H_5O-$COCH(CH_3)O$-CO, CH≡CCH_2O-CO,

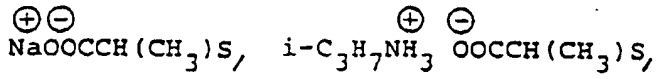




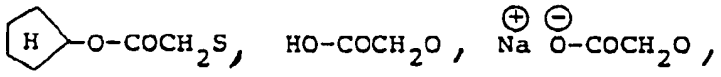
and CH_3O-CO-OCH_2S-CO;

f) X is Cl, R^1 is CH_3, R^2 is CHF_2S, R^3 is Cl and R^4 is selected from CH_2=$CHCH_2S$, CH≡CCH_2S, $C_2H_5OCH_2CH_2S$, CH_3SCH_2S, $NCCH_2S$, C_2H_5OOCS, CH_3OOCCH_2S, $C_2H_5OOCCH_2S$,

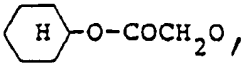
$CH_3NHOCCH_2S$, $(CH_3)_2NOCCH_2S$, $HOOCCH(CH_3)S$,



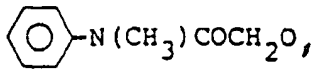
$C_2H_5OOCCH(CH_3)S$, n-$C_3H_7OOCCH(CH_3)S$, i-$C_3H_7OOCCH_2S$, $(CH_3)_2CHCH_2OOCCH_2S$, CH_2=$CHCH_2OOCCH(CH_3)S$, $C_2H_5OOC(CH_2)_3S$, $CH_3NHOCCH(CH_3)S$, $(CH_3)_2NOCCH(CH_3)S$, CH≡CCH_2NH, CH_3SO_2NH, CH_3NHSO_2NH, CH_3OOCNH, $(C_2H_5)_2NCOS$, $NCCH_2O$, ClCH=$CHCH_2O$, CH_3OCH_2O, $C_2H_5OC_2H_4O$, CH_3OCH_2S, ClCH=$CHCH_2S$, CH_2=$C(Cl)CH_2S$,



CF_3CH_2O-$COCH_2O$, CH_2=$CHCH_2O$-$COCH_2O$,



$CH_3NHCOCH_2O$, CH_2=$CHCH_2O$-$COCH(CH_3)O$, $CH_3NHCOCH(CH_3)O$, $NCCH_2O$-$COCH_2O$, CH_3O-$COCH(CH_3)S$, $NCCH_2O$-$COCH(CH_3)S$, C_2H_5O-$CO(CH_2)_3S$,

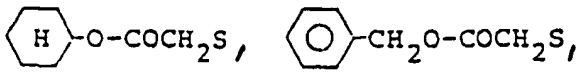


HO-$COCH_2NH$, C_2H_5O-$COCH_2NH$ and C_2H_5O-$COCH(CH_3)NH$;

g) X is F, R^1 is CH_3, R^2 is CH_3SO, R^3 is Cl and R^4 is 5-NO_2;

h) X is F, R^1 is CH_3, R^2 is CH_3S, R^3 is Cl and R^4 is selected from 5-NH_2, 5-SH, 5-$CH{\equiv}CCH_2O$, 5-$CH{\equiv}CCH_2S$ and 5-$C_2H_5OOCCH(CH_3)S$;

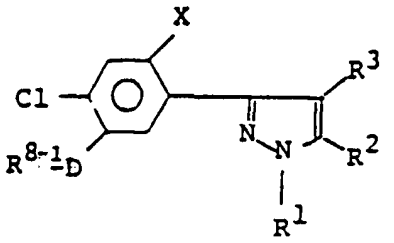
i) X is F, R^1 is CH_3, R^2 is CHF_2O, R^3 is Cl and R^4 is selected from HO-$COCH_2O$, CH_3O-$COCH_2O$, CH_2=$CHCH_2O$-$COCH_2O$, $CH{\equiv}CCH_2O$-$COCH_2O$, HO-$COCH(CH_3)O$, CH_2=$CHCH_2O$-$COCH(CH_3)O$, $CH{\equiv}CCH_2O$-$COCH(CH_3)O$, CH_2=$CHCH_2S$, CH_2=$C(CH_3)CH_2S$, $CH{\equiv}CCH_2S$, $C_2H_5OC_2H_4S$, HO-$COCH_2S$, CH_3O-$COCH_2S$, C_2H_5O-$COCH_2S$, C_2H_5O-$COCH_2S(O)$, n-C_4H_9O-$COCH_2S$, CH_2=$CHCH_2O$-$COCH_2S$, CH_3SCH_2O-$COCH_2S$,



$CH_3NHCOCH_2S$, $(CH_3)_2NCOCH_2S$, HO-$COCH(CH_3)S$, CH_3O-$COCH(CH_3)S$, C_2H_5O-$COCH(CH_3)S$, n-C_4H_9O-$COCH(CH_3)S$, CH_2=$CHCH_2O$-$COCH(CH_3)S$, $CH{\equiv}CH_2O$-$COCH(CH_3)S$, $CH_3OC_2H_4OC_2H_4O$-$COCH(CH_3)S$, CH_3SO_2NH, CH_3NHSO_2NH, C_2H_5O-$COCH_2NH$, C_2H_5O-$COCH(CH_3)NH$, CH_2=$CHCH_2O$-CO, $CH{\equiv}CCH_2O$-CO, CH_3O-$COCH(CH_3)O$-CO, C_2H_5O-$COCH(CH_3)O$-CO, $(CH_3)_2NCO$ and

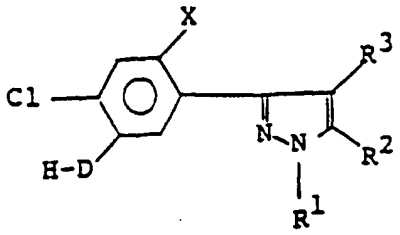
j) X is Cl, R^1 is CH_3, R^2 is CHF_2O, R^3 is Br and R^4 is $BrCH_2CH(Br)CH_2O$ or $BrC{\equiv}CCH_2O$.

3. A process for producing 3-(substituted phenyl)-pyrazole derivatives represented by the general formula



(I-2)

wherein X, R^1, R^2 and R^3 are as defined in claim 1, and D-R^{8-1} is defined as D-R^8 in claim 1 except that R^{8-1} is not hydrogen,
the process comprising reacting a pyrazole derivative represented by the general formula



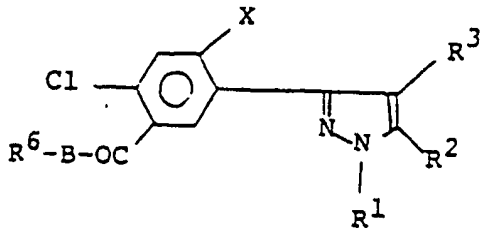
(I-1)

wherein X, R^1, R^2, R^3, and D are as detained above, with a halide represented by the general formula

$$R^{8-1}\text{-}Z \qquad \text{(II)}$$

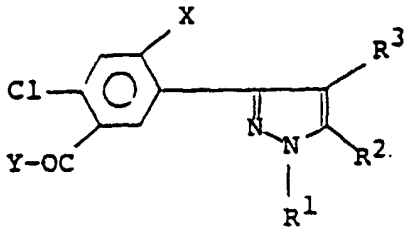
wherein R^{8-1} is defined above and Z is halogen.

4. A process for producing 3-(substituted phenyl)pyrazole derivatives represented by the general formula



(I-4)

wherein X, R^1, R^2, R^3 and -CO-B-R^6 are as defined in claim 1,
the process comprising reacting a pyrazole derivative represented by the general formula



(I-3)

wherein X, R^1, R^2 and R^3 are as defined above and Y denotes halogen or OH,
with a compound represented by the general formula

$$R^6\text{-B-H} \qquad \text{(II-1)}$$

wherein R^6 and B are as defined above.

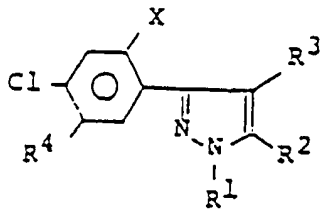
5. A herbicidal composition which contains a 3-(substituted phenyl)pyrazole derivative or a salt thereof according to claim 1 or 2 as an active ingredient.

6. Use of the herbicidal composition of claim 5, for paddy fields, upland fields, or non-farming area purposes.

7. A method for controlling undesirable plants which comprises applying a herbicidal composition containing a 3-(substituted phenyl)pyrazole derivative or a salt thereof.according to claim 1 or 2 as an active ingredient, in a dose of 1.0 g to 10 Kg in terms of the quantity of active ingredient per hectare.

Patentansprüche

1. 3-(substituiertes Phenyl)pyrazol-Derivat oder ein Salz davon, wobei das Derivat dargestellt wird durch die allgemeine Formel (I)



(I)

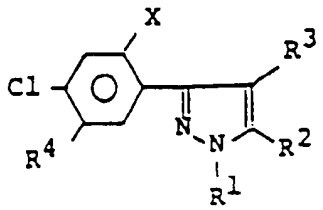
worin

X Halogen bedeutet,
R^1 CH_3 bedeutet,
R^2 Hydroxy, Mercapto, Alkylthio, Haloalkoxy oder Haloalkylthio bedeutet,
R^3 Wasserstoff oder Halogen bedeutet, und
R^4

(i) Formyl bedeutet, (ii) Nitro bedeutet,
(iii) -CO-B-R^6 bedeutet, worin B -O-, -S- oder -NR^7- bedeutet, wobei R^6 und R^7 gleich oder verschieden sind und jeweils Wasserstoff oder Cycloalkyl bedeuten, und wenn B -O- ist, R^6 ein Alkalimetallatom sein kann oder -CO-B-R^6 Methoxycarbonyl, Ethoxycarbonyl, Propoxycarbonyl, Butoxycarbonyl, Methoxycarbonylmethoxycarbonyl, Ethoxycarbonylmethoxycarbonyl, Propoxycarbonylmethoxycarbonyl, Methoxycarbonylethoxycarbonyl, Ethoxycarbonylethoxycarbonyl oder Propoxycarbonylethoxycarbonyl bedeutet,
(iv) -D-R^8 bedeutet, worin D -O-, -$S(O)_n$-, worin n eine ganze Zahl von 0 bis 2 ist, oder -NR^9- bedeutet, worin R^8 und R^9, die gleich oder verschieden sind, jeweils Wasserstoff; Alkyl; Haloalkyl; Aminosulfonyl; Phenylalkyl oder Phenoxyalkyl bedeuten, worin der Phenylring gegebenenfalls einen oder mehrere Halogensubstituenten haben kann, die gleich oder verschieden sein können; oder -D-R^8 eine Alkenyloxygruppe, enthaltend Propenyl, Butenyl oder Pentenyl; eine Alkinyloxygruppe, enthaltend Propinyl, Butinyl oder Pentinyl; eine Alkenylaminogruppe, enthaltend Propenyl, Butenyl oder Pentenyl; eine Alkoxycarbonylalkoxy- oder eine Alkylthiocarbonylalkoxygruppe, enthaltend jeweils Methyl, Ethyl, Propyl oder Butyl bedeutet; oder
(v) -$(CHR^{10})_m$-CO-E-R^{12} bedeutet, worin E -O-, -S- oder -NR^{11} bedeutet; R^{10} Wasserstoff bedeutet und R^{11} und R^{12}, die gleich oder verschieden sein können, jeweils Wasserstoff; Alkyl; Haloalkyl; Phenyl, gegebenenfalls mit einem oder mehreren Halogensubstituenten an dem Phenylring; Phenylalkyl, gegebenenfalls mit einem oder mehreren Halogensubstituenten am Phenylring, bedeutet; oder R^{11}, verbunden mit R^{12}, eine Piperidin- oder Morpholinylgruppe bedeutet; und wenn E -O- ist, R^{12} ein Alkalimetallatom sein kann; und

m eine ganze Zahl von 0 bis 3 bedeutet.

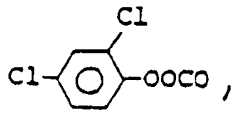
2. 3-(substituiertes Phenyl)pyrazol-Derivat oder ein Salz davon, wobei das Derivat dargestellt wird durch die allgemeine Formel (I)



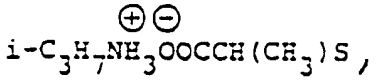
(I)

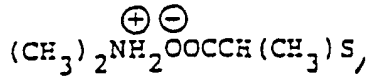
worin

a) X Cl ist, R^1 CH_3 ist, R^2 CH_3S ist, R^3 Cl ist und R^4 gewählt wird aus

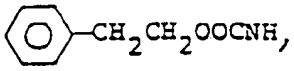


$HOOCCH(CH_3)O$, $CH_3OOCCH(CH_3)O$, $C_2H_5OOCCH(CH_3)O$, i-$C_3H_7OOCCH(CH_3)O$, CH_2=$CHCH_2OOCCH(CH_3)O$, $CH{\equiv}CCH_2OOCCH(CH_3)O$, CH_2= $CHCH_2S$, CH_2=$C(Cl)CH_2S$, $CH{\equiv}CCH_2S$, $HO_2CCH(CH_3)S$,





$Cl(CH_2)_3OOCCH(CH_3)S$, CH_2=$CHCH_2OOCCH(CH_3)S$, $CH{\equiv}CCH_2OOCCH(CH_3)S$, $C_2H_5CH(CH_3)CH_2OOCCH(CH_3)S$, $(C_2H_5)_2NCOCH(CH_3)S$, CH_3NHSO_2, $(CH_3)_2NSO_2$, n-$C_3H_7NHSO_2$, $CH_3SO_2NHSO_2$, $Cl^{\ominus}H_3N^{\oplus}$, $CH{\equiv}CCH_2NH$, CH_3OOCNH, CH_3SO_2NH, CH_3NHSO_2NH, $CH_3NHSO_2N(CH_3)$, $(CH_3)_2NSO_2N(CH_3)$, i-$C_3H_7NHSO_2NH$, $CH{\equiv}CCH_2N(CH_3)SO_2N(CH{\equiv}CCH_2)$, $CH_3OCH_2CH_2OOCNH$, $BrCH_2CH_2OOCNH$, $NCCH_2CH_2OOCNH$, CH_2=$C(CH_3)CH_2OOCNH$,



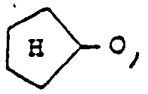
CH_2=$CHCH_2OOCCH_2NH$, $CH{\equiv}CCH_2OOCCH_2NH$, $C_2H_5OOCCH(CH_3)NH$, $HOOCCH_2NH$, CH_3OOCCH_2NH, $HOOCCH(CH_3)NH$ und $CH_3OOCCH(CH_3)NH$;

b) X Cl ist, R^1 CH_3 ist, R^2 CH_3SO ist, R^3 Cl ist und R^4 gewählt wird aus NO_2, CH_3SO_2NH, CH_3NHSO_2NH, i-$C_3H_7NHSO_2NH$ und $CH_3NHSO_2(CH{\equiv}CCH_2)$;

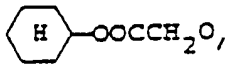
c) X Cl ist, R^1 CHF_2 ist, R^2 CH_3S ist, R^3 Cl ist und R^4 gewählt wird aus NH_2, CH_3NHSO_2NH, $CH_3NHSO_2N(CH{\equiv}CCH_2)$ und $CH_3N(CH{\equiv}CCH_2)SO_2N(CH{\equiv}CCH_2)$;

d) X Cl ist, R^1 CHF_2 ist, R^2 CH_3SO ist, R^3 Cl ist und R^4 gewählt wird aus NH_2 und NO_2;

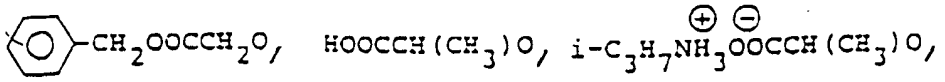
e) X Cl ist, R^1 CH_3 ist, R^2 CHF_2O ist, R^3 Cl ist und R^4 gewählt wird aus



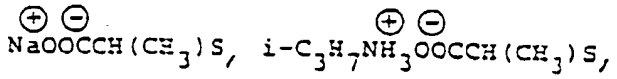
$C_2H_5OCH_2CH_2O$, $NCCH_2O$, CH_3OOCO, $n\text{-}C_6H_{13}OOCCH_2O$, $CH_2{=}CHCH_2OOCCH_2O$, $CH{\equiv}CCH_2OOCCH_2O$,



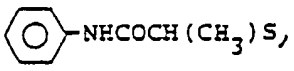
$C_2H_5O(CH_2)_2OOCCH_2O$, $C_2H_5O(CH_2)_2O(CH_2)_2OOCCH_2O$,

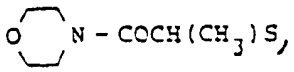


$CH_2{=}CHCH_2OOCCH(CH_3)O$, $CH{\equiv}CCH_2OOCCH(CH_3)O$, $CH_3NHCOCH(CH_3)O$, $(CH_3)_2NCOCH(CH_3)O$, $CH_2{=}CHCH_2S$, $CH_2{=}CHCH_2SO$, $CH{\equiv}CCH_2S$, $(C_2H_5O)_2P(O)CH_2S$, CH_3OOCS, CH_3OOCCH_2S, $C_2H_5OOCCH_2S$, $CH_2{=}CHCH_2OOCCH_2S$, $HOOCCH(CH_3)S$,

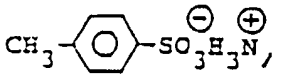


$CH_3OOCCH(CH_3)S$, $C_2H_5OOCCH(CH_3)S$, $C_2H_5OOCCH(CH_3)S(O)$, $C_2H_5OOCCH(CH_3)SO_2$, $i\text{-}C_3H_7OOCCH(CH_3)S$, $n\text{-}C_4H_9OOCCH(CH_3)S$, $(CH_3)_2CH(CH_2)_2OOCCH(CH_3)S$, $CH_2{=}CHCH_2OOCCH(CH_3)S$, $CH{\equiv}CCH_2OOCCH(CH_3)S$, $CH_3O(CH_2)_2O(CH_2)_2OOCCH(CH_3)S$, $(C_2H_5O)_2P(O)CH_2OOCCH(CH_3)S$, $CH_3NHCOCH(CH_3)S$, $(CH_3)_2NCOCH(CH_3)S$, $(C_2H_5)_2NCOCH(CH_3)S$, $i\text{-}C_3H_7NHCOCH(CH_3)S$,

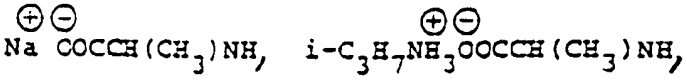




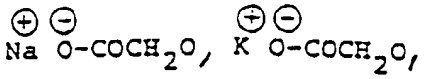
CH_3SO_2NH, CH_3NHSO_2NH, $CH_2{=}CHCH_2NH$, $CH{\equiv}CCH_2NH$, CH_3OOCNH, $Cl^{\ominus}H_3N^{\oplus}$,

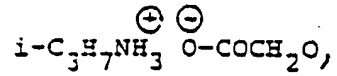


CH_3OOCCH_2NH, $HOOCCH(CH_3)NH$,

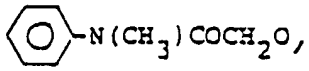


$C_2H_5OOCCH(CH_3)NH$, $CH_3NHOCCH(CH_3)NH$, $(CH_3)_2NOCCH(CH_3)NH$, CH_3NHSO_2, $(CH_3)_2NSO_2$, $(C_2H_5)_2NCOS$, $(CH_3)_3SiCH_2S$, $(CH_3)_3SiCH_2O$, $HO\text{-}COCH_2O$,

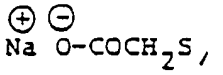


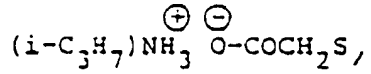


$CH_2=CHC(CH_3)_2O\text{-}COCH_2O$, $CH_3C{\equiv}CCH_2O\text{-}COCH_2O$, $CH_3NHCOCH_2O$, $(CH_3)_2NCOCH_2O$, $n\text{-}C_4H_9NHCOCH_2O$, $H_2C=CHCH_2NHCOCH_2O$, $(H_2C=CHCH_2)_2NCOCH_2O$,

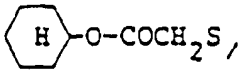


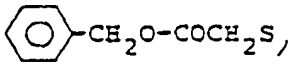
$CH_3SO_2NHCOCH_2O$, $CH_3SO_2NHCOCH(CH_3)O$, $(CH_3)_3SiCH_2O\text{-}COCH_2O$, $CH_3OC_2H_4OC_2H_4O\text{-}COCH(CH_3)O$, $CH_2=C(CH_3)CH_2S$, $CH_2=C(Cl)CH_2S$, $ClCH=CHCH_2S$, $ClCH=C(Cl)CH_2S$, $NCCH_2S$,





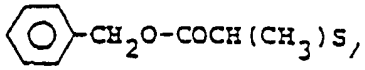
$HO\text{-}COCH_2S$, $i\text{-}C_3H_7O\text{-}COCH_2S$, $n\text{-}C_4H_9O\text{-}COCH_2S$, $C_{12}H_{25}O\text{-}COCH_2S$, $CH_3S\text{-}COCH_2S$, $C_2H_5S\text{-}COCH_2S$, $i\text{-}C_3H_7S\text{-}COCH_2S$, $CH{\equiv}CCH_2O\text{-}COCH_2S$, $C_2H_5OC_2H_4OC_2H_4O\text{-}COCH_2S$,





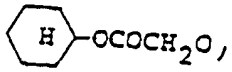
$(CH_3)_3SiCH_2O\text{-}COCH_2S$, $CH_3NHCOCH_2S$, $(CH_3)_2NCOCH_2S$, $H_2C=CHCH_2NHCOCH_2S$, $(H_2C=CHCH_2)_2NCOCH_2S$, $HC{\equiv}CCH_2NHCOCH_2S$,

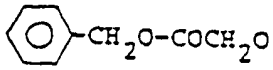
i-C_4H_9O-$COCH(CH_3)S$, t-$C_4H_9CH_2O$-$COCH(CH_3)S$, n-$C_6H_{13}O$-$COCH(CH_3)S$,



C_2H_5S-$COCH(CH_3)S$, i-C_3H_7S-$COCH(CH_3)S$, $NCCH(CH_3)O$-$COCH(CH_3)S$, $CH_3SC_2H_4O$-$COCH(CH_3)S$,

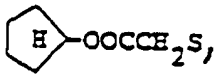
$(CH_3)_3SiCH_2O$-$COCH(CH_3)S$, CH_2=$CHCH_2NHCOCH(CH_3)S$, $(H_2C$=$CHCH_2)_2NCOCH(CH_3)S$, HC≡$CCH_2NHCOCH(CH_3)S$, CH_3CO-O-$(CH_2)_2S$, C_2H_5O-$COC(CH_3)_2S$, CH_3O-$COCH(i\text{-}C_3H_7)S$, C_2H_5O-$COCH(CH_3)N(CH_3)$, CH_2=$CHCH_2O$-CO, CH_3O-$COCH(CH_3)O$-CO, C_2H_5O-$COCH(CH_3)O$-CO, CH≡CCH_2O-CO,



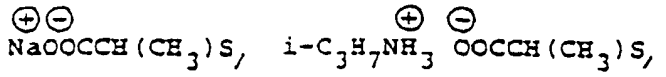


und CH_3O-CO-OCH_2S-CO;

f) X Cl ist, R^1 CH_3 ist, R^2 CHF_2S ist, R^3 Cl ist und R^4 gewählt wird aus CH_2=$CHCH_2S$, CH≡CCH_2S, $C_2H_5OCH_2CH_2S$, CH_3SCH_2S, $NCCH_2S$, C_2H_5OOCS, CH_3OOCCH_2S, $C_2H_5OOCCH_2S$,

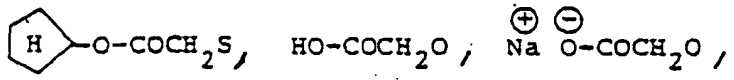


$CH_3NHOCCH_2S$, $(CH_3)_2NOCCH_2S$, HOOCCH$(CH_3)S$,

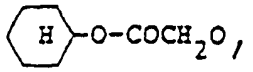


$C_2H_5OOCCH(CH_3)S$, n-$C_3H_7OOCCH(CH_3)S$, i-$C_3H_7OOCCH_2S$, $(CH_3)_2CHCH_2OOCCH_2S$, CH_2=$CHCH_2OOCCH(CH_3)S$, $C_2H_5OOC(CH_2)_3S$, $CH_3NHOCCH(CH_3)S$, $(CH_3)_2NOCCH(CH_3)S$, CH≡CCH_2NH, CH_3SO_2NH, CH_3NHSO_2NH, CH_3OOCNH, $(C_2H_5)_2NCOS$, $NCCH_2O$), ClCH=$CHCH_2O$, CH_3OCH_2O,

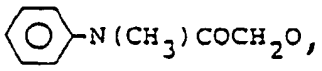
$C_2H_5OC_2H_4O$, CH_3OCH_2S, $ClCH{=}CHCH_2S$, $CH_2{=}C(Cl)CH_2S$,



$CF_3CH_2O\text{-}COCH_2O$, $CH_2{=}CHCH_2O\text{-}COCH_2O$,



$CH_3NHCOCH_2O$, $CH_2{=}CHCH_2O\text{-}COCH(CH_3)O$, $CH_3NHCOCH(CH_3)O$, $NCCH_2O\text{-}COCH_2O$, $CH_3O\text{-}COCH(CH_3)S$, $NCCH_2O\text{-}COCH(CH_3)S$, $C_2H_5O\text{-}CO(CH_2)_3S$,

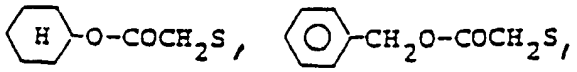


$HO\text{-}COCH_2NH$, $C_2H_5O\text{-}COCH_2NH$ und $C_2H_5O\text{-}COCH(CH_3)NH$;

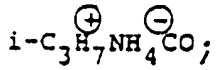
g) X F ist, R^1 CH_3 ist, R^2 CH_3SO ist, R^3 Cl ist und R^4 5-NO_2 ist;

h) X F ist, R^1 CH_3 ist, R^2 CH_3S ist, R^3 Cl ist und R^4 gewählt wird aus 5-NH_2, 5-SH, 5-$CH{\equiv}CCH_2O$, 5-$CH{\equiv}CCH_2S$ und 5-$C_2H_5OOCCH(CH_3)S$;

i) X F ist, R^1 CH_3 ist, R^2 CHF_2O ist, R^3 Cl ist und R^4 gewählt wird aus $HO\text{-}COCH_2O$, $CH_3O\text{-}COCH_2O$, $CH_2{=}CHCH_2O\text{-}COCH_2O$, $CH{\equiv}CCH_2O\text{-}COCH_2O$, $HO\text{-}COCH(CH_3)O$, $CH_2{=}CHCH_2O\text{-}COCH(CH_3)O$, $CH{\equiv}CCH_2O\text{-}COCH(CH_3)O$, $CH_2{=}CHCH_2S$, $CH_2{=}C(CH_3)CH_2S$, $CH{\equiv}CCH_2S$, $C_2H_5OC_2H_4S$, $HO\text{-}COCH_2S$, $CH_3O\text{-}COCH_2S$, $C_2H_5O\text{-}COCH_2S$, $C_2H_5O\text{-}COCH_2S(O)$, n-$C_4H_9O\text{-}COCH_2S$, $CH_2{=}CHCH_2O\text{-}COCH_2S$, $CH_3SCH_2O\text{-}COCH_2S$,

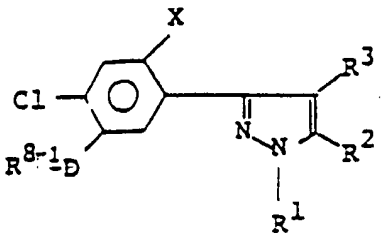


$CH_3NHCOCH_2S$, $(CH_3)_2NCOCH_2S$, $HO\text{-}COCH(CH_3)S$, $CH_3O\text{-}COCH(CH_3)S$, $C_2H_5O\text{-}COCH(CH_3)S$, n-$C_4H_9O\text{-}COCH(CH_3)S$, $CH_2{=}CHCH_2O\text{-}COCH(CH_3)S$, $CH{\equiv}CH_2O\text{-}COCH(CH_3)S$, $CH_3OC_2H_4OC_2H_4O\text{-}COCH(CH_3)S$, CH_3SO_2NH, CH_3NHSO_2NH, $C_2H_5O\text{-}COCH_2NH$, $C_2H_5O\text{-}COCH(CH_3)NH$, $CH_2{=}CHCH_2O\text{-}CO$, $CH{\equiv}CCH_2O\text{-}CO$, $CH_3O\text{-}COCH(CH_3)O\text{-}CO$, $C_2H_5O\text{-}COCH(CH_3)O\text{-}CO$, $(CH_3)_2NCO$ und



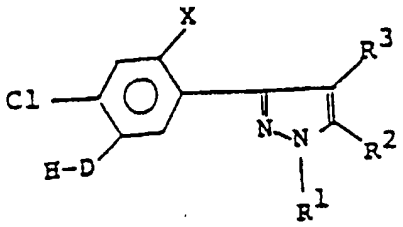
j) X Cl ist, R^1 CH_3 ist, R^2 CHF_2O ist, R^3 Br ist und R^4 $BrCH_2CH(Br)CH_2O$ oder $BrC{\equiv}CCH_2O$ ist.

3. Verfahren zur Herstellung von 3-(substituiertes Phenyl)prazol-Derivaten der allgemeinen Formel (I-2)



(I-2)

worin X, R^1, R^2 und R^3 definiert sind wie in Anspruch 1, und D-R^{8-1} definiert ist wie D-R^8 in Anspruch 1 mit dem Unterschied, daß R^{8-1} kein Wasserstoff ist,
wobei das Verfahren das Umsetzen eines Pyrazol-Derivats der allgemeinen Formel (I-1)



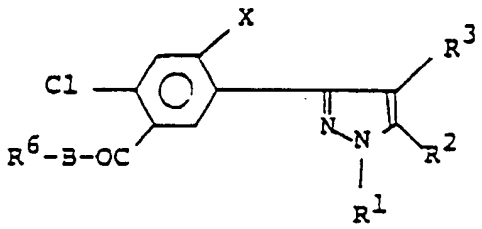
(I-1)

worin X, R^1, R^2, R^3 und D wie oben definiert sind, mit einem Halogenid der allgemeinen Formel (II)

$$R^{8-1}\text{-}Z \qquad \text{(II)}$$

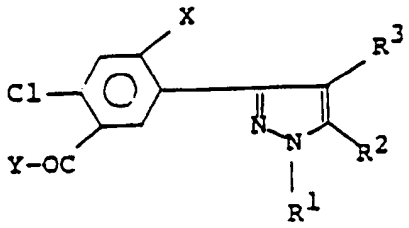
worin R^{8-1} definiert ist wie oben und Z Halogen ist, umfaßt.

4. Verfahren zur Herstellung von 3-(substituiertes Phenyl)pyrazol-Derivaten der allgemeinen Formel (I-4)



(I-4)

worin X, R^1, R^2, R^3 und -CO-B-R^6 definiert sind wie in Anspruch 1,
wobei das Verfahren das Umsetzen eines Pyrazol-Derivats der allgemeinen Formel (I-3)



(I-3)

worin X, R^1, R^2 und R^3 wie oben definiert sind und Y Halogen oder OH bedeutet, mit einer Verbindung der allge-

meinen Formel (II-1)

$$R^6\text{-B-H} \qquad \text{(II-1)}$$

worin R^6 und B wie oben definiert sind, umfaßt.

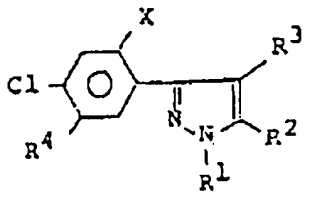
5. Herbizide Zusammensetzung, die ein 3-(substituiertes Phenyl)pyrazol-Derivat oder ein Salz davon nach Anspruch 1 oder 2 als aktiven Inhaltsstoff enthält.

6. Verwendung der herbiziden Zusammensetzung nach Anspruch 5 für Reisfelder, Hochlandfelder oder für Zwecke in nicht-landwirtschaftlichen Gebieten.

7. Verfahren zur Kontrolle von unerwünschten Pflanzen, welches das Aufbringen einer herbiziden Zusammensetzung, welche ein 3-(substituiertes Phenyl)pyrazol-Derivat oder ein Salz davon nach Anspruch 1 oder 2 als aktiven Inhaltsstoff enthält, in einer Dosis von 1,0 g bis 10 kg, bezogen auf die Menge des aktiven Inhaltsstoffs, pro Hektar umfaßt.

Revendications

1. Dérivé de 3-(phényl substitué) pyrazole ou sel de celui-ci, le dérivé étant représenté par la formule générale

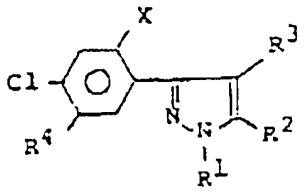


(I)

dans laquelle

X signifie un halogène,
R^1 signifie CH_3,
R^2 signifie un hydroxy, un mercapto, un alkylthio, un haloalcoxy ou un haloalkylthio,
R^3 signifie l'hydrogène ou un halogène, et
R^4 signifie (i) le formyle, (ii) le nitro, (iii) -CO-B-R^6 dans laquelle B signifie -O-, -S-, ou -NR^7, R^6 et R^7 sont les mêmes ou différents et signifient chacun l'hydrogène ou un cycloalkyle, et lorsque B est -O-, R^6 peut être un atome de métal alcalin ou -CO-B-R^6 signifie le méthoxycarbonyle, l'éthoxycarbonyle, le propoxycarbonyle, le butoxycarbonyle, le méthoxycarbonylméthoxycarbonyle, l'éthoxycarbonylméthoxycarbonyle, le propoxycarbonylméthoxycarbonyle, le méthoxycarbonyléthoxycarbonyle, l'éthoxycarbonyléthoxycarbonyle, ou le propoxycarbonyléthoxycarbonyle,
(iv) -D-R^8 dans laquelle D signifie -O-, -$S(O)_n$-, n étant un nombre entier de 0 à 2, ou -NR^9-, R^8 et R^9, qui sont les mêmes ou différents, signifient chacun l'hydrogène ; un alkyle ; un haloalyle ; un aminosulfonyle ; un phénylalkyle ou un phénoxy-alkyle ayant éventuellement sur l'anneau phényle un ou plusieurs substituants halogène qui sont les mêmes ou différents ; ou -D-R^8 signifie un groupe alcényloxy contenant le groupe propényle, buténylo ou penténylo ; un groupe alcynyloxy contenant le groupe propynyle, butynyle ou pentynyle ; un groupe alcénylamino contenant le groupe propényle, buténylo ou penténylo ; un groupe alcoxy-carbonylalcoxy ou alkylthiocarbonylalcoxy contenant chacun le méthyle, l'éthyle, le propyle ou le butyle ; ou (v) -$(CHR^{10})_m$-CO-E-R^{12} dans laquelle E signifie -O-, -S- ou -NR^{11} ; R^{10} signifie l'hydrogène et R^{11} et R^{12}, qui sont les mêmes ou différents, signifient chacun l'hydrogène ; un alkyle ; un haloalkyle ; un phényle ayant éventuellement sur l'anneau phényle un ou plusieurs substituants halogène ; un phénylalkyle ayant éventuellement sur l'anneau phényle un ou plusieurs substituants halogène ; ou R^{11}, conjointement avec R^{12} forme un groupe pipéridino ou morpholinyle, et lorsque E est -O-, R^{12} peut être un atome de métal alcalin ; et m signifie un nombre entier de 0 à 3.

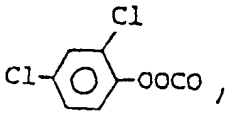
2. Dérivé de 3-(phényl substitué) pyrazole ou sel de celui-ci, le dérivé étant représenté par la formule générale



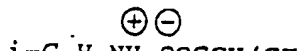
(I)

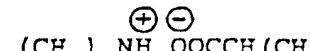
dans laquelle

a) X est Cl, R^1 est CH_3, , R^2 est CH_3S, R^3 est Cl et R^4 est sélectionné parmi

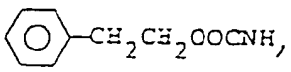


$HOOCCH(CH_3)O$, $CH_3OOCCH(CH_3)O$, $C_2H_5OOCCH(CH_3)O$, i-$C_3H_7OOCCH(CH_3)O$, $CH_2=CHCH_2OOCCH(CH_3)O$, $CH\equiv CCH_2OOCCH(CH_3)O$, $CH_2=CHCH_2S$, $CH_2=C(Cl)CH_2S$, $CH\equiv CCH_2S$, $HO_2CCH(CH_3)S$,





$Cl(CH_2)_3OOCCH(CH_3)S$, $CH_2=CHCH_2OOCCH(CH_3)S$, $CH\equiv CCH_2OOCCH(CH_3)S$, $C_2H_5CH(CH_3)CH_2OOCCH(CH_3)S$, $(C_2H_5)_2NCOCH(CH_3)S$, CH_3NHSO_2, $(CH_3)_2NSO_2$, n-$C_3H_7NHSO_2$, $CH_3SO_2NHSO_2$, $Cl^{\ominus}H_3N^{\oplus}$, $CH\equiv CCH_2NH$, CH_3OOCNH, CH_3SO_2NH, CH_3NHSO_2NH, $CH_3NHSO_2N(CH_3)$, $(CH_3)_2NSO_2N(CH_3)$, i-$C_3H_7NHSO_2NH$, $CH\equiv CCH_2N(CH_3)SO_2N(CH\equiv CCH_2)$, $CH_3OCH_2CH_2OOCNH$, $BrCH_2CH_2OOCNH$, $NCCH_2CH_2OOCNH$, $CH_2=C(CH_3)CH_2OOCNH$,



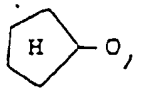
$CH_2=CHCH_2OOCCH_2NH$, $CH\equiv CCH_2OOCCH_2NH$, $C_2H_5OOCCH(CH_3)NH$, $HOOCCH_2NH$, CH_3OOCCH_2NH, $HOOCCH(CH_3)NH$ et $CH_3OOCCH(CH_3)NH$;

b) X est Cl, R^1 est CH_3, R^2 est CH_3SO, R^3 est Cl et R^4 est sélectionné parmi NO_2, CH_3SO_2NH, CH_3NHSO_2NH, i-$C_3H_7NHSO_2NH$, et $CH_3NHSO_2(CH\equiv CCH_2)$;

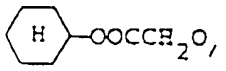
c) X est Cl, R^1 est CHF_2, R^2 est CH_3S, R^3 est Cl et R^4 est sélectionné parmi NH_2, CH_3NHSO_2NH, $CH_3NHSO_2N(CH\equiv CCH_2)$, et $CH_3N(CH\equiv CCH_2)SO_2N(CH\equiv CCH_2)$;

d) X est Cl, R^1 est CHF_2, R^2 est CH_3SO, R^3 est Cl et R^4 est sélectionné parmi NH_2 et NO_2 ;

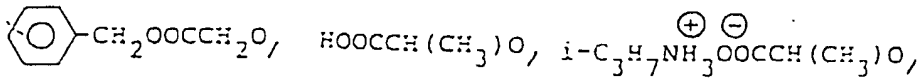
e) X est Cl, R^1 est CH_3, R^2 est CHF_2O, R^3 est Cl et R^4 est sélectionné parmi



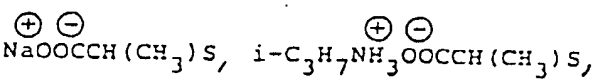
$C_2H_5OCH_2CH_2O$, $NCCH_2O$, CH_3OOCO, $n\text{-}C_6H_{13}OOCCH_2O$, $CH_2{=}CHCH_2OOCCH_2O$, $CH{\equiv}CCH_2OOCCH_2O$,



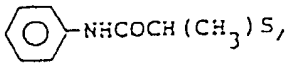
$C_2H_5O(CH_2)_2OOCCH_2O$, $C_2H_5O(CH_2)_2O(CH_2)_2OOCCH_2O$,

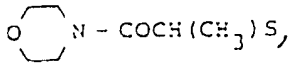


$CH_2{=}CHCH_2OOCCH(CH_3)O$, $CH{\equiv}CCH_2OOCCH(CH_3)O$, $CH_3NHCOCH(CH_3)O$, $(CH_3)_2NCOCH(CH_3)O$, $CH_2{=}CHCH_2S$, $CH_2{=}CHCH_2SO$, $CH{\equiv}CCH_2S$, $(C_2H_5O)_2P(O)CH_2S$, CH_3OOCS, CH_3OOCCH_2S, $C_2H_5OOCCH_2S$, $CH_2{=}CHCH_2OOCCH_2S$, $HOOCCH(CH_3)S$,

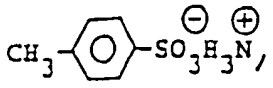


$CH_3OOCCH(CH_3)S$, $C_2H_5OOCCH(CH_3)S$, $C_2H_5OOCCH(CH_3)S(O)$, $C_2H_5OOCCH(CH_3)SO_2$, $i\text{-}C_3H_7OOCCH(CH_3)S$, $n\text{-}C_4H_9OOCCH(CH_3)S$, $(CH_3)_2CH(CH_2)_2OOCCH(CH_3)S$, $CH_2{=}CHCH_2OOCCH(CH_3)S$, $CH{\equiv}CCH_2OOCCH(CH_3)S$, $CH_3O(CH_2)_2O(CH_2)_2OOCCH(CH_3)S$, $(C_2H_5O)_2P(O)CH_2OOCCH(CH_3)S$, $CH_3NHCOCH(CH_3)S$, $(CH_3)_2NCOCH(CH_3)S$, $(C_2H_5)_2NCOCH(CH_3)S$, $i\text{-}C_3H_7NHCOCH(CH_3)S$,

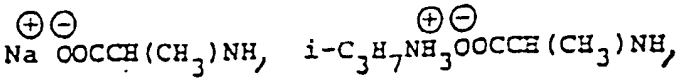




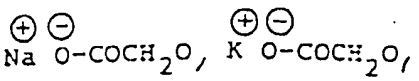
CH_3SO_2NH, CH_3NHSO_2NH, $CH_2{=}CHCH_2NH$, $CH{\equiv}CCH_2NH$, CH_3OOCNH, $Cl^{\ominus}H_3N^{\oplus}$,

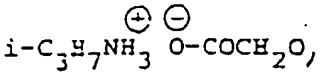


CH_3OOCCH_2NH, $HOOCCH(CH_3)NH$,

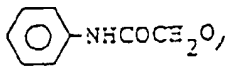


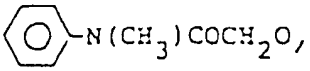
$C_2H_5OOCCH(CH_3)N$, $CH_3NHOCCH(CH_3)NH$, $(CH_3)_2NOCCH(CH_3)NH$, CH_3NHSO_2, $(CH_3)_2NSO_2$, $(C_2H_5)_2NCOS$, $(CH_3)_3SiCH_2S$, $(CH_3)_3SiCH_2O$, $HO\text{-}COCH_2O$,



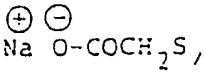


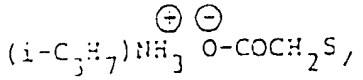
$CH_2=CHC(CH_3)_2O\text{-}COCH_2O$, $CH_3C\equiv CCH_2O\text{-}COCH_2O$, $CH_3NHCOCH_2O$, $(CH_3)_2NCOCH_2O$, n-$C_4H_9NHCOCH_2O$, $H_2C=CHCH_2NHCOCH_2O$, $(H_2C=CHCH_2)_2NCOCH_2O$,



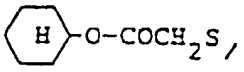


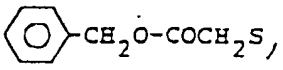
$CH_3SO_2NHCOCH_2O$, $CH_3SO_2NHCOCH(CH_3)O$, $(CH_3)_3SiCH_2O\text{-}COCH_2O$, $CH_3OC_2H_4OC_2H_4O\text{-}COCH(CH_3)O$, $CH_2=C(CH_3)CH_2S$, $CH_2=C(Cl)CH_2S$, $ClCH=CHCH_2S$, $ClCH=C(Cl)CH_2S$, $NCCH_2S$,



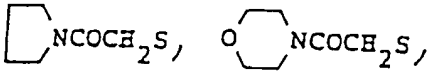


$HO\text{-}COCH_2S$, i-$C_3H_7O\text{-}COCH_2S$, n-$C_4H_9O\text{-}COCH_2S$, $C_{12}H_{25}O\text{-}COCH_2S$, $CH_3S\text{-}COCH_2S$, $C_2H_5S\text{-}COCH_2S$, i-$C_3H_7S\text{-}COCH_2S$, $CH\equiv CCH_2O\text{-}COCH_2S$, $C_2H_5OC_2H_4OC_2H_4O\text{-}COCH_2S$,

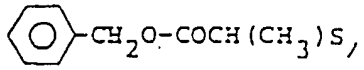




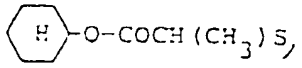
$(CH_3)_3SiCH_2O\text{-}COCH_2S$, $CH_3NHCOCH_2S$, $(CH_3)_2NCOCH_2S$, $H_2C=CHCH_2NHCOCH_2S$, $(H_2C=CHCH_2)_2NCOCH_2S$, $HC\equiv CCH_2NHCOCH_2S$,

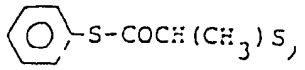


i-C_4H_9O-$COCH(CH_3)S$, t-$C_4H_9CH_2O$-$COCH(CH_3)S$, n-$C_6H_{13}O$-$COCH(CH_3)S$,

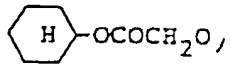


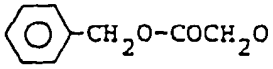
C_2H_5S-$COCH(CH_3)S$, i-C_3H_7S-$COCH(CH_3)S$, $NCCH(CH_3)O$-$COCH(CH_3)S$, $CH_3SC_2H_4O$-$COCH(CH_3)S$,





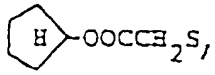
$(CH_3)_3SiCH_2O$-$COCH(CH_3)S$, $CH_2=CHCH_2NHCOCH(CH_3)S$, $(H_2C=CHCH_2)_2NCOCH(CH_3)S$, $HC\equiv CCH_2NHCOCH(CH_3)S$, CH_3CO-O-$(CH_2)_2S$, C_2H_5O-$COC(CH_3)_2S$, CH_3O-$COCH(i\text{-}C_3H_7)S$, C_2H_5O-$COCH(CH_3)N(CH_3)$, $CH_2=CHCH_2O$-CO, CH_3O-$COCH(CH_3)O$-CO, C_2H_5O-$COCH(CH_3)O$-CO, $CH\equiv CCH_2O$-CO,



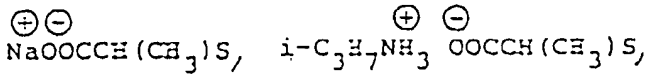


et CH_3O-CO-OCH_2S-CO;

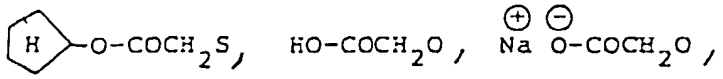
f) X est Cl, R^1 est CH_3, R^2 est CHF_2S, R^3 est Cl et R^4 est sélectionné parmi $CH_2=CHCH_2S$, $CH\equiv CCH_2S$, $C_2H_5OCH_2CH_2S$, CH_3SCH_2S, $NCCH_2S$, C_2H_5OOCS, CH_3OOCCH_2S, $C_2H_5OOCCH_2S$,



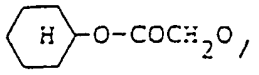
$CH_3NHOCCH_2S$, $(CH_3)_2NOCCH_2S$, $HOOCCH(CH_3)S$,



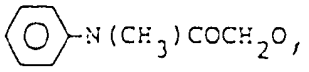
$C_2H_5OOCCH(CH_3)S$, n-$C_3H_7OOCCH(CH_3)S$, i-$C_3H_7OOCCH_2S$, $(CH_3)_2CHCH_2OOCCH_2S$, $CH_2=CHCH_2OOCCH(CH_3)S$, $C_2H_5OOC(CH_2)_3S$, $CH_3NHOCCH(CH_3)S$, $(CH_3)_2NOCCH(CH_3)S$, $CH\equiv CCH_2NH$, CH_3SO_2NH, CH_3NHSO_2NH, CH_3OOCNH, $(C_2H_5)_2NCOS$, $NCCH_2O$), $ClCH=CHCH_2O$, CH_3OCH_2O, $C_2H_5OC_2H_4O$, CH_3OCH_2S, $ClCH=CHCH_2S$, $CH_2=C(Cl)CH_2S$,



CF_3CH_2O-$COCH_2O$, CH_2=$CHCH_2O$-$COCH_2O$,



$CH_3NHCOCH_2O$, CH_2=$CHCH_2O$-$COCH(CH_3)O$, $CH_3NHCOCH(CH_3)O$, $NCCH_2O$-$COCH_2O$, CH_3O-$COCH(CH_3)S$, $NCCH_2O$-$COCH(CH_3)S$, C_2H_5O-$CO(CH_2)_3S$,

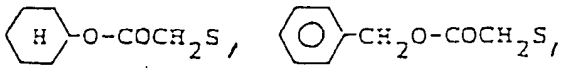


HO-$COCH_2NH$, C_2H_5O-$COCH_2NH$ et C_2H_5O-$COCH(CH_3)NH$;

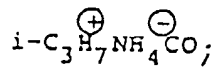
g) X est F, R^1 est CH_3, R^2 est CH_3SO, R^3 est Cl et R^4 est 5-NO_2 ;

h) X est F, R^1 est CH_3, R^2 est CH_3S, R^3 est Cl et R^4 est sélectionné parmi 5-NH_2, 5-SH, 5-CH≡CCH_2O, 5-CH≡-CCH_2S et 5-$C_2H_5OOCCH(CH_3)S$;

i) X est F, R^1 est CH_3, R^2 est CHF_2O, R^3 est Cl et R^4 est sélectionné parmi HO-$COCH_2O$, CH_3O-$COCH_2O$, CH_2-$CHCH_2O$-$COCH_2O$, CH≡CCH_2O-$COCH_2O$, HO-$COCH(CH_3)O$, CH_2=$CHCH_2O$-$COCH(CH_3)O$, CH≡CCH_2O-$COCH(CH_3)O$, CH_2=$CHCH_2S$, CH_2=$C(CH_3)CH_2S$, CH≡CCH_2S, $C_2H_5OC_2H_4S$, HO-$COCH_2S$, CH_3O-$COCH_2S$, C_2H_5O-$COCH_2S$, C_2H_5O-$COCH_2S(O)$, n-C_4H_9O-$COCH_2S$, CH_2=$CHCH_2O$-$COCH_2S$, CH_3SCH_2O-$COCH_2S$,

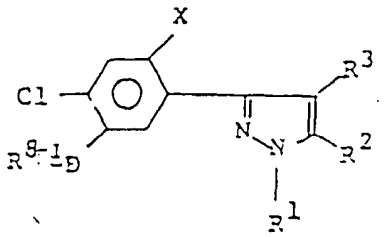


$CH_3NHCOCH_2S$, $(CH_3)_2NCOCH_2S$, HO-$COCH(CH_3)S$, CH_3O-$COCH(CH_3)S$, C_2H_5O-$COCH(CH_3)S$, n-C_4H_9O-$COCH(CH_3)S$, CH_2=$CHCH_2O$-$COCH(CH_3)S$, CH≡CH_2O-$COCH(CH_3)S$, $CH_3OC_2H_4OC_2H_4O$-$COCH(CH_3)S$, CH_3SO_2NH, CH_3NHSO_2NH, C_2H_5O-$COCH_2NH$, C_2H_5O-$COCH(CH_3)NH$, CH_2=$CHCH_2O$-CO, CH≡CCH_2O-CO, CH_3O-$COCH(CH_3)O$-CO, C_2H_5O-$COCH(CH_3)O$-CO, $(CH_3)_2NCO$ et



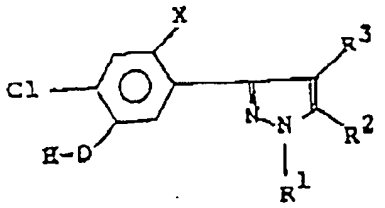
j) X est Cl, R^1 est CH_3, R^2 est CHF_2O, R^3 est Br et R^4 est $BrCH_2CH(Br)CH_2O$ ou $BrC{\equiv}CCH_2O$.

3. Procédé de production de dérivés de 3-(phényl substitué) pyrazole représentés par la formule générale



(I-2)

dans laquelle X, R^1, R^2 et R^3 sont tels que définis dans la revendication 1, et D-R^{8-1} est défini comme D-R^8 en revendication 1 sauf que R^{8-1} n'est pas l'hydrogène, le procédé comprenant la réaction d'un dérivé de pyrazole représenté par la formule générale



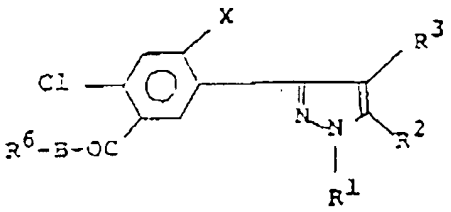
(I-1)

dans laquelle X, R^1, R^2, R^3 et D sont tels que définis ci-dessus, avec un halogènure représenté par la formule générale

$$R^{8-1}\text{-}Z \qquad \text{(II)}$$

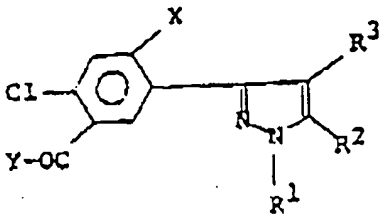
dans laquelle R^{8-1} est défini ci-dessus et Z est un halogène.

4. Procédé de production de dérivés de 3-(phényl substitué) pyrazole représentés par la formule générale



(I-4)

dans laquelle X, R^1, R^2, R^3 et -CO-B-R^6 sont tels que définis dans la revendication 1,
le procédé comprenant la réaction d'un dérivé de pyrazole représenté par la formule générale



(I-3)

dans laquelle X, R^1, R^2 et R^3 sont tels que définis ci-dessus et Y signifie un halogène ou OH,
avec un composé représenté par la formule générale

$$R^6\text{-B-H} \qquad \text{(II-1)}$$

dans laquelle R^6 et B sont tels que définis ci-dessus.

5. Composition herbicide qui contient un dérivé de 3-(phényl substitué) pyrazole ou un sel de celui-ci selon la revendication 1 ou la revendication 2 comme ingrédient actif.

6. Utilisation de la composition herbicide de la revendication 5 pour l'application à des rizières, des champs de hautes terres ou des zones non agricoles.

7. Procédé pour réguler des plantes indésirables qui comprend l'application d'une composition herbicide contenant un dérivé de 3-(phényl substitué)pyrazole ou un sel de celui-ci selon la revendication 1 ou la revendication 2 comme ingrédient actif, selon une dose de 1,0 g à 10 kg en termes de quantité d'ingrédient actif par hectare.